Table 1. Laboratory QC information for Acadia sample runs, 2002-2005.

Analysis date	QCS, 2.0 ng/L	MS (matrix spike) (+5.0 ng/L)	MS (matrix spike) (+10 ng/l)	Laboratory replicates, % difference
07/30/2002	2.1		11.6	6.2
07/30/2002	2.1		11.6	6.2
07/30/2002	2.1		11.6	6.2
07/30/2002	2.1	5.3	11.1	2.2
07/30/2002	2.1	5.3	11.1	2.2
07/08/2003	2.0	6.2		0.2
07/15/2003	2.0	6.2		7.3
07/30/2003	2.0	5.8		4.3
07/30/2003	2.0	6.5		5.8
11/30/2003	2.0	6.2		3.7
11/30/2003	2.0	5.6		0.2
01/25/2004	2.0	5.9		1.3
02/27/2004	2.0	5.8		0.2
02/27/2004	2.0	7.2		1.4
03/19/2004	2.0	5.4		2.3
06/10/2004	2.0	5.7		5.1
06/10/2004	2.0	6.1		0.3
09/09/2004	2.0	5.7		5.1
09/09/2004	2.0	5.3		5.3
09/29/2004	2.0		16.3	1.1
09/29/2004	2.0		17.4	1.6
10/19/2004	2.0	5.2		2.4
10/19/2004	2.0	5.2		2.4
10/22/2004	2.0		21.0	1.7
10/22/2004	2.0		17.4	0.4
11/02/2004	2.0		17.4	1.9
11/02/2004	2.0		17.2	1.6
11/02/2004	2.0		17.4	1.9
11/02/2004	2.0		17.2	1.6
11/06/2004	2.0		16.1	2.8
11/06/2004	2.0		18.8	0.8
11/16/2004	2.0		23.9	3.2
11/16/2004	2.0		24.3	4.4
12/03/2004	1.9	5.9		1.1
12/03/2004	1.9	6.2		1.5
12/09/2004	2.0	5.3		4.8
12/14/2004	1.9	6.4		1.6
12/14/2004	1.9	9.5		5.4
01/19/2005	2.0		16.3	1.5
01/19/2005	2.0		18.6	3.1
01/28/2005	1.9		12.5	0.3
02/02/2005	2.0	5.7		8.9
02/02/2005	2.0	5.3		9.7
02/10/2005	2.1		16.4	1.2
02/10/2005	2.1		13.2	0.2

02/17/2005	2.0		20.0	2.9
02/17/2005	2.0		13.2	1.2
02/28/2005	2.0		12.0	1.5
02/28/2005	2.1		12.8	2.7
03/05/2005	2.1		27.5	3.3
03/05/2005	2.1		13.1	13.4
03/11/2005	2.0		17.1	3.7
04/27/2005	2.0		18.9	0.7
04/27/2005	2.0		14.6	0.8
04/27/2005	2.0		18.9	0.7
04/29/2005	2.0	5.5		3.6
04/29/2005	2.0	5.9		7.4
04/29/2005	2.0	5.9		7.4

Table 2. Field replicates taken during 2002-2005 at Acadia National Park.

Sample ID	Sample Date	Regular sample value (ng/L)	Duplicate sample value (ng/L)	% difference
HB1	05/15/2002	1.47	1.85	-11.4
CB1	06/19/2002	0.31	0.36	-7.5
HB1	07/06/2002	1.05	1.02	1.4
HB1	07/21/2004	0.83	0.78	3.1
HB1	09/15/2004	0.80	0.91	-6.4
CB1*	10/06/2004	0.85	1.53	-28.6
EIEN**	10/06/2004	0.70	2.36	-54.2
HB1	11/03/2004	1.66	1.86	-5.7
CB1	12/01/2004	0.37	1.86	-66.8
HB1	02/09/2005	0.52	0.80	-21.2
HT1E-S	03/05/2005	6.64	6.83	-1.4
CB1	03/10/2005	0.52	0.47	5.1
CB1	04/05/2005	0.69	0.64	3.8
CB1	05/22/2005	0.66	0.46	17.9
HB1	05/29/2005	0.98	0.96	1.0
CB1	01/24/2002	0.37	0.34	4.2
CB1	02/20/2002	0.17	0.16	3.0
CB1	03/13/2002	0.22	0.24	-4.3
CB1	04/04/2002	0.56	0.55	0.9
CB1	05/02/2002	0.51	0.62	-9.7
CB1	05/22/2002	0.46	0.66	-17.9
CB1	06/19/2002	0.31	0.36	-7.5
HB1	01/09/2002	0.44	0.32	15.8
HB1	02/06/2002	0.43	0.40	3.6
HB1	03/06/2002	0.64	0.64	0.0
HB1	03/27/2002	4.22	4.87	-7.2
HB1	04/24/2002	0.52	0.63	-9.6
HB1	05/29/2002	0.96	0.98	-1.0
CB1	08/04/2004	0.43	0.31	16.2

* Debris noted in stream; many leaves in sample pool.

** We believe this was a labeling error, and the duplicate was taken at EIES, which had a value of 0.93 ng/L, yielding a 14% difference.

TF Collection Procedure - Plastic

1. Get equipment out of pack: 1 plastic bottle (hereafter 'new bottle'), poly wool, beaker, field sheets, labels, forceps, sharpie.
2. Unscrew overflow bottle if it has water in it. Pour water into plastic beaker & record total volume of water that was in overflow. Replace overflow bottle (no rinsing necessary)
3. Unscrew sample bottle that's in the collector (hereafter 'old bottle') from lid & leave it in the soda bottle holder.
4. Uncap new bottle, place new bottle's cap on old bottle.
5. Immediately label old bottle – must have site name (e.g., C1A) & Date. If label won't stick, write on the lid or where label-residue is stuck.
6. Remove used wool from funnel & discard. Remove any rough debris (twigs) from funnel – only things that won't fit down the tube.
7. Rinse funnel with the new bottle's DI and pour all the new DI in to rinse interior of whole apparatus (funnel, tubing). Lift end of tube up & down to get lid & make sure it flows all the way through.
   ** **For Equipment Blanks (1 per watershed per collection)** **
   After rinsing with the new bottle (350 mL) of water, pour the full bottle's water (there will be 1 packed per trip, labeled 'DIW for Blank') through the apparatus & into the new bottle. Use the 'DIW for Blank' bottle as the new bottle in the collector. Label the full blank 'EQBLK-Site' (e.g. – EQBLK-C1A), Date.
8. Screw on new bottle and replace poly wool with fresh wool – just a tiny bit – we are just keeping needles & bugs from going down the tube! Use forceps to push down into funnel neck (just a bit – to keep it from blowing out – don't shove all the way into funnel neck!)

Notes:

- Keep good field notes, as always. Be sure to write down if wool was missing when you got to the site.
- Label samples immediately – there are no identifying marks on bottles.
- If tubing starts to look gooey, clean gently (cleaning procedure on other pages). Note cleaning date on field sheet.
- Inspect apparatus before re-setting – rodents sometimes chew tubing. You have spare tubing to replace it with if this happens; just make a note in the book.

TF Collection Procedure - Mercury

1. Get equipment out of pack: 1 glass bottle (hereafter 'new bottle'), 2 gloves, glass wool, sharpie.
2. Put on gloves.
3. Open Ziplocs on new bottle so you can get to it easily.
4. Unscrew sample bottle that's in the collector (hereafter 'old bottle') from lid & leave it in the soda bottle holder.
5. Uncap new bottle, place new bottle's cap on old bottle.
6. Remove new bottle from plastic bags & set aside (carefully, for a moment). Put old bottle in the bags, immediately label it by writing site name (e.g., C1A) directly on the outer bag at least twice.
7. Remove used wool from funnel & discard (careful, this is glass fibers!) Remove any rough debris (twigs) from funnel with gloved finger – only things that won't fit down the tube.
8. Rinse funnel with the new bottle's DI and pour all the new DI in to rinse interior of whole apparatus (funnel, tubing). Lift end of tube up & down to get lid & make sure it flows all the way through.
   **** For Equipment Blanks (1 per watershed per collection) ****
   Do not let all the water run all the way through – lift the lid up to stop the flow of the last ~250 mL of water (keep in funnel for a moment) – then have a 250 mL Teflon bottle ready and catch that last 250 mL in the Teflon bottle. Label it 'EQBLK-Site' (e.g. – EQBLK-C1A), Date.
9. Screw on new bottle and replace glass wool with fresh wool – just a tiny bit – we are just keeping needles & bugs from going down the tube!

Notes for Hg:

- Keep as clean as possible. Don't scratch head, hair, etc. Don't breathe all over the inside of the apparatus.
- Careful – the glass is very slippery with gloves on. The glass wool will get in your skin (like fiberglass). Handle with gloves.
- Keep good field notes, as always.
- Label samples immediately – there are no identifying marks on bottles.
- If a bottle breaks, bag it with many bags & hike out with it secured far from your body (outer pocket of pack).
- Do not kink the Teflon tubing – it does not re-form.
- If tubing starts to look gooey, clean gently (cleaning procedure on other pages). Note cleaning date on field sheet.

Appendix C. Analytical Methods for the University of Maine Laboratory.

Analyte	*Method*	*Reference*
pH, closed cell	Electrode	Hillman *et al.*[6], EPA 19.0[5]
pH, aerated	Electrode	Hillman *et al.*[6], EPA 5.0[5]
Specific conductance	Wheatstone bridge	EPA 120.1[2],EPA 23.0[5]
True color	Spectrophotometer, 457.5 nm	EPA 110.2[2]
ANC	Gran Titration	Hillman *et al.*[6], EPA 5.0[5]
Anions: Cl, NO_3, SO_4	Ion chromatography	EPA 300.0[1]
Pre 1999 analysis methods for Ca, Mg, Na, K, and Al		
Calcium	AAS with N_20-acetylene flame	EPA 215.1[2]
Magnesium	AAS with N_20-acetylene flame	EPA 242.1[2]
Sodium	AAS with air-acetylene flame	EPA 258.1[2]
Potassium	AAS with air-acetylene flame	EPA 273.1[2]
Aluminum (total)	AAS with graphite furnace	EPA 200.9[1]
1999 to 2003 methods for Ca, Mg, Na, K, and Al		
Calcium	Inductively Coupled Atomic Emission Spectroscopy (ICP)	EPA 200.15[10]
Magnesium	Inductively Coupled Atomic Emission Spectroscopy (ICP)	EPA 200.15[10]
Sodium	Inductively Coupled Atomic Emission Spectroscopy (ICP)	EPA 200.15[10]
Potassium	Inductively Coupled Atomic Emission Spectroscopy (ICP)	EPA 200.15[10]
Aluminum (total)	Inductively Coupled Atomic Emission Spectroscopy (ICP)	EPA 200.15[10]
2003 to present methods for Ca, Mg, Na, K, NH_4		
Calcium	Ion Chromatography (IC)	ASTM D 6919-03[11]
Magnesium	Ion Chromatography (IC)	ASTM D 6919-03[11]
Sodium	Ion Chromatography (IC)	ASTM D 6919-03[11]
Potassium	Ion Chromatography (IC)	ASTM D 6919-03[11]
Ammonium	Ion Chromatography (IC)	ASTM D 6919-03[11]
2003 to present methods for Al		
Aluminum (total)	AAS with graphite furnace	EPA 200.9[1]
Dissolved Organic Carbon	IR C analyzer, persulfate oxidation	EPA 415.1[2]
Ammonium (prior to 2003)	Autoanalyzer	EPA 9.0[5] and Bran & Luebbe 780-86T[7]

Silica	Autoanalyzer	EPA 22.0[5] and Bran & Luebbe 785-86T[7]
Total Hg	CVAFS dual gold trap	EPA 1631[8]
MethylHg	Distillation, Aqueous Ethylation, Purge and Trap, and CVAFS	EPA 1630[9]

AAS=atomic absorption spectrophotometry IR=Infrared Spectrophotometry
CVAFS=cold vapor atomic fluorescence spectrometry

Method references:

1. Methods for the Determination of Inorganic Substances in Environmental Samples, EPA 600/R-93-100, 1993.
2. Methods for Chemical Analysis of Water and Wastes, EPA 600/4-79-020, 1979, Revised 1983.
3. Standard Methods for Examination of Water and Wastewater, 18th ed. 1992.
4. Methods for the Determination of Metals in Environmental Samples, EPA 600/4-91/010, 1991, Supplement 1, EPA 600/R-94/111, 1994.
5. Handbook of Methods for Acid Deposition Studies: Laboratory Analysis For Surface Water Chemistry, EPA 600/4-87-026, 1987.
6. Hillman,D.C., J. Potter, and S. Simon, 1986. Analytical methods for the National Surface Water Survey, Eastern Lake Survey. EPA/600/4-86/009, EPA Las Vegas.
7. Bran & Luebbe Manual
8. Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry, EPA 821-R-99-005, 1999
9. Methyl Mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry, EPA Draft, 1998
10 EPA 200.15, Determination of Metals and Trace Elements in Water by Ultrasonic Nebulization Inductively Coupled Plasma-Atomic Emission Spectrometry. Rev. 1.2, 1994.
11 ASTM D 6919-03 Standard Test Method for Determination of Dissolved Alkali and Alkaline Earth Cations and Ammonium in Water and Wastewater by Ion Chromatography. Annual Book of ASTM Standards, Vol 11.01, 2003.

Table 1. Summary of 2001 accuracy data for QC solutions.

ANALYTE	UNITS	NOMINAL VALUE	ACTUAL VALUES	n	NET MEAN BIAS %
CLPH	SU	4.70	4.70 ± 0.01	67	0.07
EQPH	SU	4.70	4.71 ± 0.07	158	0.13
ANC	µeq/L	40	40.8 ± 1.23	73	2.04
DIC	mg/L C	5.00	5.00 ± 0.23	128	-0.07
DOC	mg/L C	5.00	4.92 ± 0.22	184	-1.66
COLOR	PCU	10.0	9.96 ± 0.30	171	-0.35
COND	µS/cm	10	9.96 ± 0.37	139	-0.36
Ca	mg/L	1.00	1.02 ± 0.05	327	1.95
Mg	mg/L	1.00	1.02 ± 0.04	327	1.58
Na	mg/L	1.00	1.01 ± 0.04	327	0.64
K	mg/L	1.00	1.01 ± 0.05	327	0.57
Cl	µeq/L	50.0	51.0 ± 2.84	107	2.08
NO3	µeq/L	25.0	25.93 ± 1.1	105	3.73
SO4	µeq/L	50.0	51.0 ± 3.16	107	2.09
Total P	µg/L	50.2	49.9 ± 1.84	96	-0.66
Total N	mg/L	0.559	0.550 ± 0.01	168	-1.62
Si	mg/L	2.00	1.97 ± 0.09	184	-1.45
NH4	mg/L	0.83	0.85 ± 0.01	56	2.09
Al	µg/L	200.0	200.1 ± 9.92	327	0.04

Table 2. Summary of 2001 data for field blanks.

Variable	Median	Range	QA Objective	n
pH, equilibrated	NA	NA	NA	NA
pH, ARAS	NA	NA	NA	NA
ANC (µeq/L)	-1.1	-48.3 to 1.4	10	13
Conductivity (µS/cm)	1.34	0.85 to 11.39	4	13
DOC (mg/L)	NA	NA	NA	NA
DIC (mg/L)	NA	NA	NA	NA
Al, total dissolved (µg/L)	0.3	0 to 3.6	10	11
Ca (mg/L)	0.01	0 to 0.03	0.02	13
Mg (mg/L)	0.00	0 to 0.01	0.02	13
Na (mg/L)	0.01	0 to 0.03	0.02	13
K (mg/L)	0.01	0 to 0.03	0.02	13
NH_4 (mg/L)	0.02	0 to 0.02	0.05	12
Cl (µeq/L)	1.4	0.9 to 4.0	1.0	13
NO_3 (µeq/L)	0	0 to 0.3	0.2	13
SO_4 (µeq/L)	0.5	0 to 2.3	2.0	13
Total P	0	0 to 0.7	1.0	3
Total N (mg/L)	0.001	0 to 0.024	0.050	13
Si (mg/L)	0	0 to 0.06	0.1	12
Apparent Color (PCU)	0.5	0 to 1	0	10
Chla	NA	NA	NA	NA

Table 3. Itemization of 2001 sample blank QC violations defined as two times the MDL.

Date	K (mg/L)	Na (mg/L)	Cl (µeq/L)	NO_3 (µeq/L)	DOC (mg/L)	Si (mg/L)	Total P (µg/L)
1/2/01		0.03	2.0		0.30		
1/28/01					0.50		
2/13/01			1.1		0.50		
3/7/01				0.30			
3/8/01				0.40			
3/15/01						0.18	
4/3/01						0.18	
4/10/01			1.3		0.30		
4/12/01			1.1				1.3
4/15/01			1.1		0.40		
4/17/01							1.3
4/20/01					0.30		
4/23/01			1.1				
4/26/01			1.3				
4/29/01					0.40		
5/1/01		0.05	1.8				
5/15/01					0.30		
5/29/01				0.30			
6/1/01					0.30		
6/12/01					0.30		
6/26/01			1.9				
7/15/01			1.8			0.19	
7/17/01			2.1				
7/24/01			1.9				
8/22/01	0.03		1.1				
8/28/01			1.3				
9/7/01			1.9				
9/11/01			1.1				
9/18/01			1.1				
9/24/01	0.03	0.03	2.0				
10/16/01	0.03		1.5				
10/29/01			1.3				
11/17/01			1.5				
11/26/01			2.2				
12/3/01		0.04	2.4		0.47		
12/12/01			1.9				
12/26/01	0.03	0.04	2.9		0.34		

NOTE: DOC values in red represent those samples filtered through a glass 0.7 µm filter. Values in black Represent those samples filtered through a 0.4 µm filter.

Table 4. Summary of 2001 precision data for field replicates.

Variable	Median	Range	QA Objective	n
pH, closed cell (values ≤ 5.74)	0.01	-0.5 to 0.1	± 0.075	26
pH, closed cell (values > 5.74)	-0.01	-0.14 to 0.15	± 0.15	34
pH, equilibrated (values ≤ 5.74)	-0.01	-0.26 to 0.04	± 0.075	15
pH, equilibrated (values > 5.74)	-0.01	-0.22 to 0.06	± 0.15	46
ANC (values ≤100 µeq/L)	0.1	-4.2 to 4.5	± 5 µeq/L	53
ANC (values >100 µeq/L)	0.37%	0 to 0.90%	± 5%	10
Conductivity (values ≤ 50uS/cm)	0	-1.7 to 2.3	± 1 uS/cm	60
Conductivity (values > 50uS/cm)	2.23%	0.26 to 4.2%	± 2%	2
DOC (values ≤ 2 mg/L)	0	-0.5 to 0.14	± 0.1 mg/L	19
DOC (values > 2 mg/L)	1.57%	0 to 25.4%	± 5%	42
DIC (values ≤ 2 mg/L)	0	-0.3 to 0.6	± 0.1 mg/L	42
DIC (values > 2 mg/L)	2.77%	1.89 to 10.3%	± 5%	7
Al, total dissolved (values ≤ 100 µg/L)	-1.7	-28.7 to 12.7	± 10 µg/L	34
Al, total dissolved (values > 100 µg/L)	0.60%	0 to 12.2%	± 10%	22
Ca (values ≤ 0.4 mg/L)	0	0 to 0	± 0.02 mg/L	1
Ca (values > 0.4 mg/L)	1.02%	0 to 4.12%	± 5%	61
Mg (values ≤ 0.2 mg/L)	0	0 to 0	± 0.01 mg/L	2
Mg (values > 0.2 mg/L)	0.18%	0 to 3.07%	± 5%	60
Na (mg/L)	1.15%	0 to 12.0%	± 5%	62
K (values ≤ 0.8 mg/L)	0	-0.14 to 0.1	± 0.04 mg/L	57
K (values > 0.8 mg/L)	1.55%	0 to 7.04%	± 5%	5
NH_4 (mg/L)	0	-0.03 to 0.02	± 0.02 mg/L	62
Cl (values ≤ 20 µeq/L)	0.15	-1.6 to 2.4	± 1.0 µeq/L	6
Cl (values > 20 µeq/L)	0.68%	0 to 10.0%	± 5%	56
NO_3 (values ≤ 10 µeq/L)	0	-0.3 to 0.4	± 0.5 µeq/L	52
NO_3 (values > 10 µeq/L)	0.37%	0 to 12.5%	± 5%	10
SO_4 (ueq/L)	0.36%	0 to 106.6%	± 5%	62
Total P (values ≤ 20 µg/l)	0	-5 to 3	± 1 µg/L	58
Total P (values > 20 µg/l)	6.17%	5.2 to 7.1%	± 5%	2
Total N (values ≤ 0.020 mg/l)	0.001	0 to 0.001	± 0.001	2
Total N (values > 0.020 mg/l)	2.57%	0 to 41.3%	± 5%	59
Si (values ≤ 1 mg/L)	0	-0.05 to 0.04	± 0.05 mg/L	14
Si (values > 1 mg/L)	1.72%	0 to 10.5%	± 5%	48
True Color (values ≤ 50 PCU)	0	-1 to 1	± 5 PCU	56
True Color (values > 50 PCU)	1.09%	0.4 to 42.6%	± 10%	4

Table 5. Itemization of 2001 field replicate precision violations.

Site	Sample Date	Analyte	Duplicate Values	Precision
WB3	1-2-01	DOC	1.3, 1.5	-0.20 mg/L
WB3	1-2-01	Total P	4.3, 2.5	1.80 µg/L
EB8	1-16-01	EqpH	5.84, 6.06	-0.22
EB8	1-16-01	Total P	1.8, 3.4	-1.6 µg/L
EB8	1-16-01	Total N	0.06, 0.07	11.79%
WB3	2-13-01	DOC	1.2, 1.7	-0.50 mg/L
EB8	4-3-01	DIC	0.7, 0.9	-0.20 mg/L
EB8	4-3-01	DOC	2.3, 1.6	25.38%
EB8	4-3-01	Total P	8.6, 12.8	-4.20 µg/L
EB8	4-3-01	Total N	0.061, 0.045	21.35%
WB3	4-17-01	DIC	0.6, 0.4	0.20 mg/L
WB3	4-17-01	DOC	2.1, 3.0	24.96%
EBT-0	5-1-01	SO_4	88.9, 12.5	106.55%
EBT-0	5-1-01	EqpH	5.6, 5.86	-0.26
EBT-0	5-1-01	Total P	2, 3.4	-1.40 µg/L
EB8	5-1-01	Total N	0.064, 0.069	5.32%
EB8	5-29-01	ClpH	5.7, 5.6	0.10
EB8	5-29-01	DOC	1.8, 2.3	-0.50 mg/L
EB8	5-29-01	Total N	0.079, 0.070	8.54%
WB3	6-5-01	Total N	0.241, 0.279	10.33%
WB3	12-26-01	DIC	0.41, 0.6	-0.19 mg/L
WB3	12-26-01	Sp. Cond.	43.5, 41.2	2.30 µS/cm
Sinclair	7-17-01	Total P	14, 19	-5.00 µg/L
Sinclair	7-17-01	Color	121, 65	-42.58%
HB1	1-2-01	Alx	42.8, 61.6	-18.80 µg/L
HB1	1-2-01	AlOrg	54, 37.1	16.90 µg/L
CB1	2-27-01	Al	76.9, 64.2	12.7 µg/L
CB1	2-27-01	Alx	57.3, 44.1	13.20 µg/L
HB1	3-13-01	K	0.18, 0.23	-0.05 mg/L
HB1	3-13-01	NO_3	10.8, 12.9	12.3%
HB1	4-10-01	AlOrg	63.9, 80.1	-16.20 µg/L
CB1	5-29-01	Al	43.5, 72.2	-28.7 µg/L
CB1	5-29-01	Total N	0.035, 0.032	6.33%
CB1	6-28-01	DIC	2.2, 1.9	10.35%
CB1	6-28-01	Total N	0.063, 0.115	41.31%
HB1	7-18-01	K	0.17, 0.31	-0.14 mg/L
HB1	7-18-01	DOC	1.6, 1.9	-0.30 mg/L
HB1	7-18-01	Total P	2.3, 3.4	-1.10 µg/L
HB1	7-18-01	Total N	0.239, 0.385	33.09%
HB1	10-19-01	Alx	22.5, 43.8	-21.3 µg/L
HB1	10-19-01	AlOrg	57.3, 38.5	18.80 µg/L
CB1	11-17-01	K	0.18, 0.11	0.07 mg/L
CB1	11-17-01	ClpH	5.72, 6.22	-0.50
CB1	11-17-01	DIC	1.75, 1.64	0.11 mg/L
CB1	11-17-01	DOC	0.95, 0.81	0.14 mg/L
CB1	11-17-01	Total N	0.078, 0.05	30.94%
CB1	12-12-01	Total N	0.032, 0.038	12.12%
Loon	9-29-01	Total P	9.3, 12	-2.7 µg/L
PINEO	10-21-01	Cl	66.9, 61	6.52%
PINEO	10-21-01	DOC	4.93, 4.4	8.03%
Gould	10-28-01	K	1.05, 1.16	7.04%
Gould	10-28-01	DOC	4.01, 4.49	7.99%
Gould	10-28-01	Total P	14, 11	3.0 µg/L
Rocky	10-31-01	Total P	5.1, 3.9	1.2 µg/L
Bracey	11-14-01	Total P	7, 8.2	-1.2 µg/L
LARDP	11-17-01	DOC	3.03, 3.37	7.51%
ROUNL	11-18-01	Na	1.08, 1.28	11.98%
Wiley	3-8-01	Na	0.99, 1.07	5.49%
Wiley	3-8-01	K	0.24, 0.29	-0.1 mg/L
Wiley	3-8-01	Cl	20.3, 23.4	10.03%
Mud O	3-15-01	EqpH	4.77, 4.83	-0.1
Newbert O	3-28-01	Alx	5, 17	-12.0 µg/L
Newbert O	3-28-01	Sp. Cond.	51.78, 54.95	4.20 µS/cm
Jellison	4-12-01	K	0.25, 0.35	-0.1 mg/L
Abol	4-19-01	Cl	14.9, 16.5	-1.6 µeq/L
Second	5-8-01	Total P	5.9, 8	-2.1 µg/L
Second	5-8-01	Sp. Cond.	22.23, 23.9	-1.7 µS/cm
Bean O	5-7-01	ClpH	5.96, 5.81	0.2
Bean O	5-7-01	DIC	2, 1.4	0.6 mg/L
Abol	5-16-01	Total P	21.7, 19.7	7.11%
Salmon E	7-25-01	Total P	7.9, 5.8	2.1 µg/L
Abol E	7-31-01	DOC	2.8, 2.6	5.24%
Abol E	7-31-01	Total P	6.6, 4.2	2.4 µg/L
Crystal E	8-4-01	ClpH	5.46, 5.37	0.1
Crystal E	8-4-01	Total P	8.3, 6.6	1.7 µg/L
Bean E	8-7-01	Cl	13.7, 11.3	2.4 µeq/L
Bean E	8-7-01	DIC	0.6, 0.9	-0.3 mg/L
Bracey	10-16-01	DIC	1.8, 1.6	0.2 mg/L
Bracey	10-16-01	Si	0.57, 0.62	-0.1 mg/L
Partridge	10-16-01	Cl	44, 40.1	6.56%
Partridge	10-16-01	DIC	0.86, 0.66	0.2 µg/L
Partridge	10-16-01	DOC	2.74, 2.39	9.65%
Wizard	10-16-01	Total P	6.1, 9.1	-3.0 µg/L
Russell Pond	7-25-01	Total P	2.4, 4.3	-1.9 µg/L
Scokes	8-21-01	Total P	42, 39	5.24%
Somers	8-22-01	Al	52.1, 63.1	-11.0 µg/L
Somers	8-22-01	Cl	40.3, 36.9	6.23 µeq/L
Somers	8-22-01	Si	1.74, 1.5	10.48%

Table 6. Summary of 2001 precision data for laboratory duplicates.

Variable	Median	Range	QA Objective	n
pH, closed cell (values ≤ 5.74)	-0.01	-0.06 to 0.21	± 0.075	21
pH, closed cell (values > 5.74)	0.01	-0.08 to 1.00	± 0.15	11
pH, equilibrated (values ≤ 5.74)	0	-0.12 to 0.03	± 0.075	33
pH, equilibrated (values > 5.74)	-0.01	-0.06 to 0.03	± 0.15	18
ANC (values ≤100 µeq/L)	0.15	-1.9 to 2.00	± 5 µeq/L	32
ANC (values >100 µeq/L)	0.53%	0.36 to 0.70%	± 5%	2
Conductivity (values ≤ 50uS/cm)	-0.10	-0.84 to 1.14	± 1 uS/cm	50
Conductivity (values > 50uS/cm)	0.79%	0.79 to 0.79%	± 2%	1
DOC (values ≤ 2 mg/L)	0.01	-0.02 to 0.02	± 0.1 mg/L	9
DOC (values > 2 mg/L)	0.52%	0 to 6.75%	± 5%	26
DIC (values ≤ 2 mg/L)	0	-0.05 to 0.06	± 0.1 mg/L	17
DIC (values > 2 mg/L)	0.95%	0 to 1.9%	± 5%	2
Al, total dissolved (values ≤ 100 µg/L)	0.1	-2.5 to 3.8	± 10 µg/L	17
Al, total dissolved (values > 100 µg/L)	0.56%	0 to 1.74%	± 10%	18
Ca (values ≤ 0.4 mg/L)	0	-0.01 to 0	± 0.02 mg/L	14
Ca (values > 0.4 mg/L)	1.24%	0 to 6.72%	± 5%	35
Mg (values ≤ 0.2 mg/L)	0	0 to 0	± 0.01 mg/L	2
Mg (values > 0.2 mg/L)	0%	0 to 6.64%	± 5%	36
Na (values ≤ 0.46 mg/L)	0	-0.01 to 0.01	± 0.02	13
Na (values > 0.46 mg/L)	0.88%	0 to 3.85%	± 5%	41
K (values ≤ 0.8 mg/L)	0	-0.01 to 0.03	± 0.04 mg/L	45
K (values > 0.8 mg/L)	0.86%	0.86 to 0.86%	± 5%	1
NH_4 (values ≤ 0.36 mg/L)	0	-0.07 to 0.18	± 0.02 mg/L	90
NH_4 (values > 0.36 mg/L)	1.63%	0 to 3.79%	± 5%	6
Cl (values ≤ 20 µeq/L)	0	-0.34 to 0.29	± 1.0 µeq/L	16
Cl (values > 20 µeq/L)	0.48%	0.02 to 5.64%	± 5%	25
NO_3 (values ≤ 10 µeq/L)	0	-0.14 to 0.19	± 0.5 µeq/L	31
NO_3 (values > 10 µeq/L)	0.31%	0 to 1.39%	± 5%	10
SO_4 (values ≤ 20 µeq/L)	0.08	-0.17 to 0.34	± 1.0 µeq/L	6
SO_4 (values > 20 µeq/L)	0.16%	0.01 to 1.81%	± 5%	35
Total P (values ≤ 20 µg/l)	0	-1.50 to 0.60	± 1 µg/L	11
Total P (values > 20 µg/l)	3.4%	2.16 to 14.45%	± 5%	3
Total N (mg/l)	1.24%	0 to 13.47%	± 5%	77
Si (values ≤ 1 mg/L)	0	-0.07 to 0.15	± 0.05 mg/L	59
Si (values > 1 mg/L)	0.64%	0 to 21.9%	± 5%	43
True Color (values ≤ 50 PCU)	0	-1.00 to 1.00	± 5 PCU	45
True Color (values > 50 PCU)	0%	0 to 0%	± 10%	1

Table 1. Summary of 2002 accuracy data for QC solutions.

ANALYTE	UNITS	NOMINAL VALUE	ACTUAL VALUES	n	NET MEAN BIAS %
CLPH	SU	4.70	4.70 ± 0.02	96	0.07
EQPH	SU	4.70	4.70 ± 0.01	216	0.07
ANC	µeq/L	40	39.6 ± 1.91	96	-1.02
DIC	mg/L C	5.00	5.05 ± 0.16	107	0.98
DOC	mg/L C	5.00	5.00 ± 0.14	226	0.00
COLOR	PCU	10.0	10.08 ± 0.51	196	0.77
COND (Jan to Mar)	µS/cm	10	9.69 ± 0.18	13	-3.15
COND (Apr to Dec)	µS/cm	23.8	24.03 ± 0.55	110	0.97
Ca	mg/L	1.00	0.98 ± 0.05	272	-2.00
Mg	mg/L	1.00	1.00 ± 0.05	272	-0.18
Na	mg/L	1.00	1.00 ± 0.05	272	0.25
K	mg/L	1.00	0.99 ± 0.05	272	-1.06
Cl	µeq/L	50.0	48.5 ± 1.91	214	-2.97
NO3	µeq/L	25.0	24.72 ± 0.80	214	-1.13
SO4	µeq/L	50.0	48.7 ± 1.76	214	-2.54
Total P	µg/L	57.2	56.9 ± 3.25	110	-0.47
Total N	mg/L	0.559	0.546 ± 0.01	117	-2.38
Si	mg/L	2.09	2.09 ± 0.06	134	0.20
NH4	mg/L	0.54	0.55 ± 0.02	111	2.37
Al	µg/L	200.0	199.1 ± 8.54	278	-0.44

Table 2. Summary of data for field blanks.

Variable	Median	Range	QA Objective	n
PH, equilibrated	NA	NA	NA	NA
PH, ARAS	NA	NA	NA	NA
ANC (µeq/L)	0	-13 to 6	10	11
Conductivity (µS/cm)	1.71	0.58 to 2.3	4	11
DOC (mg/L)	0.38	0.38 to 0.38	0.2	1
DIC (mg/L)	NA	NA	NA	NA
Al, total dissolved (µg/L)	1.40	0 to 3.4	10	11
Ca (mg/L)	0.02	0 to 0.03	0.02	11
Mg (mg/L)	0.00	0 to 0	0.02	11
Na (mg/L)	0.01	0 to 0.05	0.02	11
K (mg/L)	0.02	0.01 to 0.05	0.02	11
NH_4 (mg/L)	0.01	0 to 0.03	0.05	11
Cl (µeq/L)	1.6	0.4 to 3.2	1.0	11
NO_3 (µeq/L)	0	0 to 0.2	0.2	11
SO_4 (µeq/L)	0.7	0 to 1.4	2.0	11
Total P	NA	NA	NA	NA
Total N (mg/L)	0.009	0 to 0.024	0.050	11
Si (mg/L)	0	0 to 0.03	0.1	11
Apparent Color (PCU)	0	0 to 4	0	11
Chla	NA	NA	NA	NA

Table 3. Itemization of sample blank QC violations defined as two times the MDL.

Date	Ca (mg/L)	K (mg/L)	Na (mg/L)	Cl (µeq/L)	NO_3 (µeq/L)	Al Org (µg/L)	DOC (mg/L)
1/9/02				2.5			
1/24/02				1.4			
2/6/02				1.4			0.33
2/13/02		0.03		1.5			0.30
2/20/02				1.4			
2/28/02				1.3			0.28
3/5/02		0.03		1.3			
3/13/02		0.03		1.1			
3/17/02				1.3			
3/20/02		0.03		1.8			
3/27/02		0.03		1.5			0.25
4/5/02							0.32
4/17/02		0.03		1.6			0.25
5/14/02		0.03					
5/19/02					0.2		0.22
5/22/02		0.03					
5/29/02		0.03			0.2		
6/6/02		0.03					
6/11/02		0.03					0.33
7/6/02				1.5			0.29
7/24/02				1.2			
7/30/02				1.7			
8/1/02				1.7			
8/6/02				1.2			
8/8/02				1.1			
8/14/02	0.03	0.03		1.2			
8/20/02	0.03			1.7			0.23
8/27/02	0.03			1.2			0.25
8/28/02							0.32
9/2/02	0.03	0.03		1.2		16.1	0.23
9/8/02		0.03		1.7			
9/18/02		0.03	0.04				
9/24/02				1.1			0.29
9/26/02		0.03	0.04	2.1			0.28
9/30/02		0.03	0.03	1.1			
10/1/02							0.31
10/9/02				1.4			
10/15/02							0.24
10/21/02				1.1			
10/28/02		0.03	0.04	1.5	0.4		0.25
10/30/02		0.03		1.5			0.24
11/12/02							0.25
11/17/02							0.35
11/23/02							0.74
11/26/02							0.79
12/11/02				1.6			
12/17/02				1.2			0.66
12/22/02				1.5			
12/27/02							

NOTE: DOC values in red represent those samples filtered through a glass 0.7 µm filter. Values in black Represent those samples filtered through a 0.4 µm filter.

Table 4. Summary of precision data for field replicates.

Variable	Median	Range	QA Objective	n
pH, closed cell (values ≤ 5.74)	0.01	-0.09 to .17	± 0.075	31
pH, closed cell (values > 5.74)	0.01	-0.12 to 0.15	± 0.15	35
pH, equilibrated (values ≤ 5.74)	-0.01	-0-04 to 0.01	± 0.075	21
pH, equilibrated (values > 5.74)	-0.01	-0.18 to 0.07	± 0.15	45
ANC (values ≤100 µeq/L)	0.1	-26.9 to 3.7	± 5 µeq/L	59
ANC (values >100 µeq/L)	0.65%	0 to 2.05%	± 5%	7
Conductivity (values ≤ 50uS/cm)	0	-1.6 to 1.3	± 1 uS/cm	61
Conductivity (values > 50uS/cm)	0.47%	0.24 to 0.60%	± 2%	5
DOC (values ≤ 2 mg/L)	0.02	-0.13 to 0.16	± 0.1 mg/L	19
DOC (values > 2 mg/L)	1.59%	0 to 19.9%	± 5%	47
DIC (values ≤ 2 mg/L)	0	-2.4 to 0.2	± 0.1 mg/L	57
DIC (values > 2 mg/L)	2.89%	1.26 to 4.16%	± 5%	5
Al, total dissolved (values ≤ 100 µg/L)	0.2	-6.2 to 6.9	± 10 µg/L	25
Al, total dissolved (values > 100 µg/L)	1.18%	0 to 6.2%	± 10%	32
Ca (values ≤ 0.4 mg/L)	0.01	0.01 to 0.01	± 0.02 mg/L	1
Ca (values > 0.4 mg/L)	0.99%	0 to 11.9%	± 5%	65
Mg (values ≤ 0.2 mg/L)	0.01	0 to 0.01	± 0.01 mg/L	2
Mg (values > 0.2 mg/L)	0.50%	0 to 10.4%	± 5%	64
Na (mg/L)	1.43%	0 to 15.0%	± 5%	66
K (values ≤ 0.8 mg/L)	0	-0.08 to 0.11	± 0.04 mg/L	62
K (values > 0.8 mg/L)	2.74%	1.64 to 4.36%	± 5%	4
NH_4 (mg/L)	0	-0.02 to 0.02	± 0.02 mg/L	65
Cl (values ≤ 20 µeq/L)	0	-2 to 8.5	± 1.0 µeq/L	10
Cl (values > 20 µeq/L)	0.63%	0 to 6.7%	± 5%	56
NO_3 (values ≤ 10 µeq/L)	0	-0.6 to 0.3	± 0.5 µeq/L	58
NO_3 (values > 10 µeq/L)	0.26%	-0.2 to 0.54%	± 5%	8
SO_4 (ueq/L)	0%	0 to 2.9%	± 5%	66
Total P (µg/l)	0.3	-3 to 7	± 1 µg/L	33
Total N (mg/l)	2.58%	0 to 68%	± 5%	42
Si (values ≤ 1 mg/L)	0	-0.07 to 0.05	± 0.05 mg/L	21
Si (values > 1 mg/L)	0%	0 to 23%	± 5%	45
True Color (values ≤ 50 PCU)	0	-3 to 2	± 5 PCU	58
True Color (values > 50 PCU)	0%	0 to 0%	± 10%	5

Table 5. Itemization of field replicate precision violations.

Site	Sample Date	Analyte	Duplicate Values	Precision
Gould	10-23-02	DOC	5.26, 4.65	8.71%
Gould	10-23-02	Total P	17, 15	2.00 µg/L
Gould	10-23-02	Total N	0.35, 0.378	5.44%
Black	10-27-02	DIC	2, 1.8	0.20 mg/L
Black	10-27-02	Total P	10, 7.1	2.90 µg/L
SANDB	11-16-02	Ca	1, 0.93	5.13%
SANDB	11-16-02	DOC	4.29, 3.99	5.12%
SANDB	11-16-02	Total P	17, 10	7.00 µg/L
Hamil	11-20-02	Total N	0.25, 0.182	22.26%
Rock Pond	10-9-02	Na	0.88, 0.75	11.28%
Rock Pond	10-9-02	K	0.4, 0.33	0.07 mg/L
Rock Pond	10-9-02	Cl	13, 10	3.00 µeq/L
Greenwood Pd	10-9-02	Ca	0.5, 0.43	10.64%
Greenwood Pd	10-9-02	DOC	1.89, 2.11	7.78%
CB1	1-24-02	Ca	0.89, 0.96	5.35%
CB1	1-24-02	Mg	0.35, 0.38	5.81%
CB1	2-20-02	Total N	0.023, 0.028	13.86%
HB1	3-6-02	ClpH	5.48, 5.57	-0.09
CB1	3-13-02	ClpH	5.66, 5.57	0.09
CB1	3-13-02	DIC	0.7, 0.9	-0.02 mg/L
CB1	3-13-02	Total N	0.07, 0.053	19.55%
HB1	4-24-02	Total P	2.5, 1.1	1.40 µg/L
HB1	5-29-02	ANC	41.1, 68	-26.90 µeq/L
HB1	5-29-02	Ca	1.47, 1.74	11.90%
HB1	5-29-02	Mg	0.44, 0.51	10.42%
HB1	5-29-02	K	0.25, 0.3	-0.05 mg/L
HB1	5-29-02	EqpH	6.83, 7.01	-0.18
HB1	5-29-02	Si	2.8, 3.9	23.22%
HB1	5-29-02	Total N	0.054, 0.153	67.64%
CB1	6-19-02	Total N	0.077, 0.098	16.97%
HB1	7-6-02	Ca	1.89, 2.05	5.74%
HB1	7-6-02	Mg	0.55, 0.6	6.15%
HB1	7-6-02	DIC	1.1, 3.5	-2.40 mg/L
HB1	7-6-02	Total P	2.7, 4.2	-1.50 µg/L
HB1	7-6-02	Total N	0.12, 0.168	23.57%
Salmon E	11-4-02	DIC	1.6, 1.4	0.20 mg/L
Newbert E	7-24-02	Si	0.86, 0.93	-0.07 mg/L
Abol E	7-31-02	DOC	2.08, 2.76	19.87%
Abol	10-15-02	Cl	20, 22	6.73%
Bean E	8-1-02	Cl	12, 14	-2.00 µeq/L
Bean E	8-1-02	DIC	0.9, 0.7	0.20 mg/L
Bracey E	5-16-02	K	0.3, 0.24	0.06 mg/L
Crystal E	8-5-02	ClpH	5.44, 5.27	0.17
Abol O	4-18-02	DIC	1.7, 2	-0.30 mg/L
Abol O	4-18-02	Total P	9.5, 5.2	4.30 µg/L
Bean O	4-30-02	Cl	19, 16.6	2.40 µeq/L
Bean O	4-30-02	Total P	13, 9.7	20.56%
Bean O	5-6-02	Total N	0.235, 0.262	7.68%
Mud O	3-4-02	K	0.41, 0.36	0.05 mg/L
Mud O	3-4-02	DIC	0.5, 0.7	-0.20 mg/L
Mud O	3-4-02	Total N	0.169, 0.149	8.89%
Mud O	3-28-02	DOC	4.92, 4.53	5.84%
Mud O	3-28-02	Sp Cond	33.50, 35.00	-1.50 µS/cm
Partridge O	4-22-02	DIC	0.6, 0.8	-0.20 mg/L
Wiley O	3-11-02	Sp Cond	28.00, 29.60	-1.60 µS/cm
Round Pond	7-15-02	DIC	1.4, 1.6	-0.20 mg/L
Miller	9-2-02	Na	0.99, 0.8	15.01%
Miller	9-2-02	K	0.29, 0.18	0.11 mg/L
Miller	9-2-02	Cl	18, 9.5	8.50%
Miller	9-2-02	Sp Cond	20.1, 18.8	1.30 µS/cm
Round	9-2-02	DIC	1.6, 2	-0.40 mg/L
Baker	6-6-02	K	0.37, 0.45	-0.08 mg/L
Baker	6-6-02	Cl	38, 41	5.37%
Baker	6-6-02	NO_3	0, 0.6	-0.60 µeq/L
EB8	4-5-02	DOC	2.36, 2.15	6.59%
EB8	4-5-02	Total N	0.094, 0.081	10.51%
EB8	4-30-02	DOC	2.04, 1.88	5.77%
EB8	4-30-02	Total P	1.4, 4.4	-3.00 µg/L
EB8	4-30-02	Total N	0.085, 0.075	8.84%
EB8	5-28-02	DOC	2.08, 1.9	6.40%
EB8	5-28-02	Total P	4.7, 2.2	2.50 µg/L
WB3	2-5-02	Total P	3.1, 2	1.10 µg/L
WB3	2-26-02	Total P	1.1, 3	-1.90 µg/L
WB3	3-20-02	Total P	4.4, 2	2.40 µg/L
WB3	5-14-02	DOC	1.93, 1.77	0.16 mg/L
WB3	5-14-02	Total N	0.181, 0.219	13.44%
WB3	6-11-02	Total N	0.113, 0.134	12.02%

Table 6. Summary of precision data for laboratory duplicates.

Variable	Median	Range	QA Objective	n
pH, closed cell (values ≤ 5.74)	0	-0.85 to 0.07	± 0.075	44
pH, closed cell (values > 5.74)	0.01	-0.18 to 0.08	± 0.15	89
pH, equilibrated (values ≤ 5.74)	0	-0.4 to 0.04	± 0.075	57
pH, equilibrated (values > 5.74)	0	-0.53 to 0.12	± 0.15	81
ANC (values ≤100 µeq/L)	-0.09	-10.77 to 9.01	± 5 µeq/L	93
ANC (values >100 µeq/L)	0.39%	0 to 3.45%	± 5%	58
Conductivity (values ≤ 50uS/cm)	-0.04	-0.71 to 0.41	± 1 uS/cm	74
Conductivity (values > 50uS/cm)	0.20%	0 to 0.72%	± 2%	8
DOC (values ≤ 2 mg/L)	-0.02	-0.3 to 0.17	± 0.1 mg/L	15
DOC (values > 2 mg/L)	0.74%	0 to 13.9%	± 5%	85
DIC (values ≤ 2 mg/L)	0	-0.10 to 0.11	± 0.1 mg/L	50
DIC (values > 2 mg/L)	1.66%	0 to 3.9%	± 5%	10
Al, total dissolved (values ≤ 100 µg/L)	-0.03	-4.9 to 4.4	± 10 µg/L	52
Al, total dissolved (values > 100 µg/L)	0.57%	0 to 5.62%	± 10%	29
Ca (values ≤ 0.4 mg/L)	0	-0.02 to 0.005	± 0.02 mg/L	13
Ca (values > 0.4 mg/L)	0.76%	0 to 8.55%	± 5%	69
Mg (values ≤ 0.2 mg/L)	0	-0.002 to 0.001	± 0.01 mg/L	16
Mg (values > 0.2 mg/L)	0.73%	0 to 7.16%	± 5%	65
Na (values ≤ 0.46 mg/L)	0	-0.002 to 0.02	± 0.02	12
Na (values > 0.46 mg/L)	1.03%	0 to 7%	± 5%	69
K (values ≤ 0.8 mg/L)	0	-0.02 to 0.03	± 0.04 mg/L	77
K (values > 0.8 mg/L)	1.52%	0 to 7.44%	± 5%	33
NH_4 (values ≤ 0.36 mg/L)	0	-0.01 to 0.01	± 0.02 mg/L	83
NH_4 (values > 0.36 mg/L)	0%	0 to 0%	± 5%	1
Cl (values ≤ 20 µeq/L)	0.04	-0.98 to 1.35	± 1.0 µeq/L	39
Cl (values > 20 µeq/L)	0.14%	0 to 2.87%	± 5%	105
NO_3 (values ≤ 10 µeq/L)	0	-0.68 to 2.52	± 0.5 µeq/L	120
NO_3 (values > 10 µeq/L)	0.23%	0.05 to 0.83%	± 5%	24
SO_4 (values ≤ 20 µeq/L)	0	-0.23 to 0.20	± 1.0 µeq/L	14
SO_4 (values > 20 µeq/L)	0.12%	0 to 1.93%	± 5%	130
Total P (values ≤ 20 µg/l)	0.10	-4.60 to 5.80	± 1 µg/L	72
Total P (values > 20 µg/l)	5.41%	0 to 63.23%	± 5%	30
Total N (mg/l)	0.85%	0 to 8.47%	± 5%	73
Si (values ≤ 1 mg/L)	0	-0.04 to 0.05	± 0.05 mg/L	42
Si (values > 1 mg/L)	1.10%	0 to 5.83%	± 5%	42
True Color (values ≤ 50 PCU)	0	-1.00 to 1.00	± 5 PCU	98
True Color (values > 50 PCU)	0%	0 to 0%	± 10%	13

Table 1. Summary of 2003 accuracy data for QC solutions.

Analyte	Units	Standard Value	Mean Actual Value	SD	n	Net Mean Bias %
ClpH	SU	4.70	4.7	0.0	148	0.4
EqpH	SU	4.70	4.7	0.1	67	0.6
ANC STD Acid	SU	4.70	4.7	0.1	193	0.3
ANC 20	ueq/L	20	19.89	1.8	50	-0.5
ANC 40	ueq/L	40	39.36	1.7	40	-1.6
ANC 80	ueq/L	80	78.60	1.2	20	-1.8
Total ANC	ueq/L	20,40,80	na	na	110	-1.1
Sp Cond 74	uS/cm	74	73.8	1.5	54	-0.3
Sp Cond 147	uS/cm	147	144.8	3.8	40	-1.5
Sp Cond 718	uS/cm	718	715.2	9.2	25	-0.4
Total Sp Cond	uS/cm	74,147,718	na		117	-0.8
Cl 50	ueq/L	50	50.3	1.3	31	0.6
Cl 25	ueq/L	25	25.1	0.7	8	0.4
Total Cl	ueq/L	50,25	na	na	39	0.5
SO4 100	ueq/L	100	104.5	5.7	24	4.5
SO4 50	ueq/L	50	50.5	1.0	6	1.0
SO4 20	ueq/L	20	20.7	0.2	9	3.4
Total SO4	ueq/L	100,50,20	na	na	39	3.7
NO3 5	ueq/L	5	4.87	0.08	8	-2.6
NO3 10	ueq/L	10	9.71	0.27	29	-2.9
Total NO3	ueq/L	5,10	na	na	37	-2.8

Table 2. Summary of data for field blanks; only reported if above instrument detection level.

Analyte	Units	Median of differences	Range	QA Objective	n
EqpH	SU	5.70	5.62 to 5.78	5.40 to 5.90	9
ANC	ueq/L	0.84	-1.36 to 2.19	<4	7
Sp Cond	uS/cm	1.4	1.2 to 1.5	<2	9
Cl	ueq/L	0.5	0.3 to 1.5	<1	10
SO4	ueq/L	0.2	0.2 to 0.4	<2	3
NO3	ueq/L	0.11	0.11 to 0.11	<0.2	1
Color	PCU	0	0 to 2.5	<0	8

Table 3. Itemization of sample blank QC violations defined as two times the MDL. Bold-underlined values are out of range.

Analyte	Units	MDL	LBL-1	LBL-2	LBL-3	LBL-4	LBL-5	LBL-6	LBL-7	LBL-8	LBL-9	LBL-10
EqpH	SU	na	5.60	5.69	5.68	5.75	5.74	5.68	5.69	5.66	5.68	5.71
ANC	µeq/L	na	-0.87	-0.02	1.22	-2.09						
Cond	µS/cm	na	1.2	1.4	0.8	0.8	1.5	1.1	1.2	0.3		
Color	PCU	na	0	0	0	0	**2.5**	0	0	0		
Cl	µeq/L	0.8	<0.8	<0.8	<0.8	<0.8	0.8	<0.8	<0.8	<0.8	<0.8	<0.8
SO4	µeq/L	0.8	<0.8	<0.8	<0.8	<0.8	<0.8	<0.8	<0.8	<0.8	<0.8	<0.8
NO3	µeq/L	0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5

Table 4. Summary of precision data for field replicates.

Analyte	Median of differences	Range	QA Objective	n
ClpH (values ≤ 5.74)	0.00	-0.04 to 0.05	± 0.075	12
ClpH (values > 5.74)	0.00	-0.06 to 0.04	± 0.15	14
EqpH (values ≤ 5.74)	-0.01	-0.18 to 0.02	± 0.075	10
EqpH (values > 5.74)	-0.02	-0.27 to 0.06	± 0.15	16
ANC (values ≤ 100 µeq/L)	0.73	-8.97 to 5.07	± 5 µeq/L	19
ANC (values > 100 µeq/L)	0.60%	0.26 to 3.09 %	± 5%	5
Conductivity (values ≤ 50 µS/cm)	0.0	-0.8 to 1.2	± 1 µS/cm	22
Conductivity (values > 50 µS/cm)	0.1%	0.1 to 2.1 %	± 2%	3
Color (values ≤ 50 PCU)	0	-5 to 5	± 5 PCU	21
Color (values > 50 PCU)	0.0%	0.0 to 15.7 %	± 10%	4
Cl (values ≤ 20 µeq/L)	0.7	0.65 to 0.65	± 1 µeq/L	1
Cl (values > 20 µeq/L)	0.2%	0.1 to 1.4 %	± 5%	3
NO3 (values ≤ 10 µeq/L)	0.02	-0.40 to 0.30	± 0.5 µeq/L	4
SO4 (µeq/L)	1.1%	0.1 to 2.1 %	± 5%	4

Table 5. Itemization of field replicate precision violations.

Sample ID	Date	Analyte	Values	Values	Precision
Mud-O	3/24/2003	ANC	-4.28	-13.25	-8.97
Second-E	5/21/2003	Sp Cond	15.5	23.9	1.17
Round-E	7/22/2003	Sp Cond	55.5	53.9	2.1%
Round-E	7/22/2003	Anc	-99.56	-94.49	5.07
Mud-E	7/31/2003	EqpH	5.23	5.41	-0.18
Bracey-E	11/3/2003	EqpH	6.80	6.53	-0.27

Table 6. Summary of precision data for laboratory duplicates.

Analyte	Median of differences	Range	QA Objective	n
EqpH (values ≤ 5.74)	-0.01	-0.03 to 0.07	± 0.075	29
EqpH (values > 5.74)	0.37	0.00 to 3.00	± 0.15	36
ANC (values ≤ 100 µeq/L)	0.8	-8.56 to 15.21	± 5 µeq/L	48
ANC (values > 100 µeq/L)	0.43%	0.22 to 3.6 %	± 5%	4
Conductivity (values ≤ 50 µS/cm)	-0.0	-0.7 to 1.7	± 1 µS/cm	63
Conductivity (values > 50 µS/cm)	0.8%	0.6 to 1.0 %	± 2%	2
Color (values ≤ 50 PCU)	0	-5 to 2.5	± 5 PCU	48
Cl (values ≤ 20 µeq/L)	0.10	-0.1 to 0.3	± 1 µeq/L	2
Cl (values > 20 µeq/L)	0.20%	0.0 to 2.4%	± 5%	28
NO3 (values ≤ 10 µeq/L)	0	-0.59 to 0.1	± 0.5 µeq/L	18
SO4 (µeq/L)	0.19%	0.0 to 1.5 %	± 5%	30

Determination of base cations using Ion Chromatography

- We analyzed water samples for cations using IC and compared those results to ICP results to validate the decision to change analysis methods.
- We analyzed samples with low organic concentrations and samples with high organic concentrations (labeled Low and High DOC).
- We also analyzed the same samples twice on the IC to indicate result reproducibility, with different operators and calibrations.
- The black line on each graph is the 1:1 line. The colored line, equation, and r^2 value are the regression for low and high DOC samples combined

Determination of base cations using Ion Chromatography

- Each analyte has two slides associated with it:
  - Slide one:
    - The left graph represents the average of the results of the two IC runs compared to the ICP results in *µeq/L*.
    - The right graph represents the results of the two IC runs compared to each other in *µeq/L*.
  - Slide two:
    - The left graph represents the comparison of average IC results to ICP results in *mg/L*.
    - The right graph represents the comparison of AA results to ICP results in *mg/L*.
    - Notice that both the IC and AA results are higher than the ICP.

Samples Analyzed

Low DOC

BBWM

EB8	18-Nov
WB3	18-Nov
IE1	13-Nov
IE3	13-Nov
IW1	13-Nov
EB8	11-Nov
WB3	11-Nov
EB8	4-Nov
WB3	4-Nov
IE1	29-Oct
IE5	29-Oct
IE6	30-Oct
IE10	30-Oct
IWB1	29-Oct
IWB3	29-Oct
IWB5	30-Oct
IWB9	30-Oct

RLTM

Tunk-o	18-Nov
Anderson In 1	18-Nov
Anderson In 2	18-Nov
Anderson-E	18-Nov

Acadia

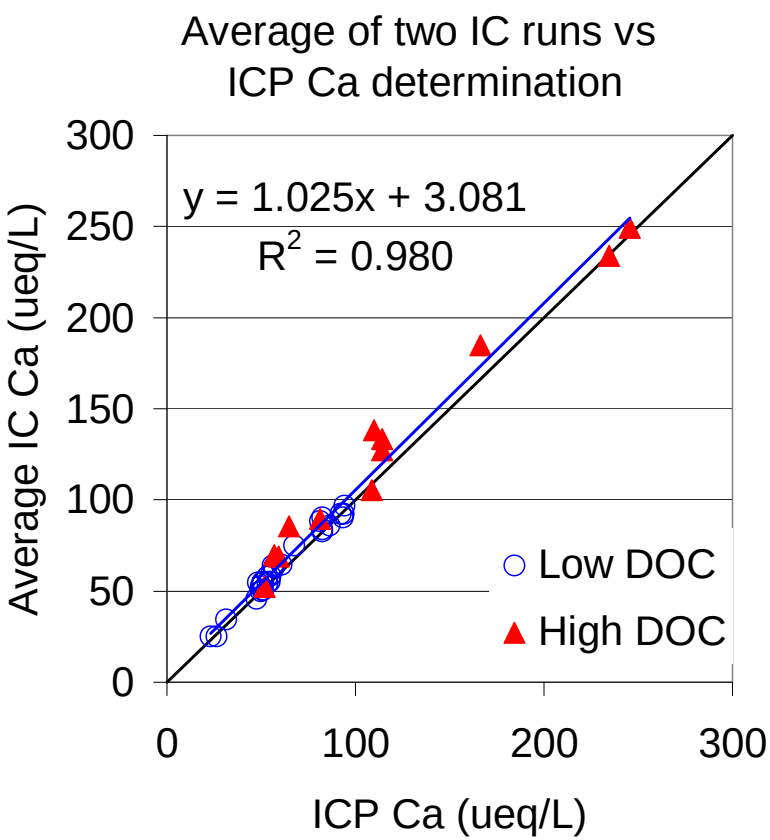
CB1	16-Nov
HB1	16-Nov
IH1	13-Nov
IH2	13-Nov
IH6	14-Nov
IH12	15-Nov
CB1	10-Nov
HB1	10-Nov

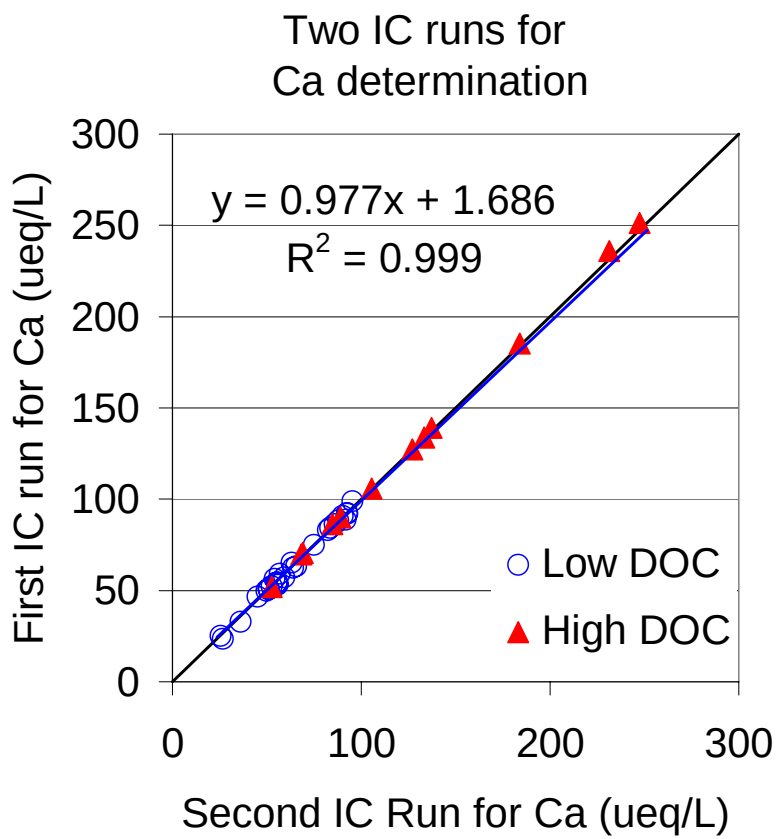
All samples takes in 2003; N=41

High DOC

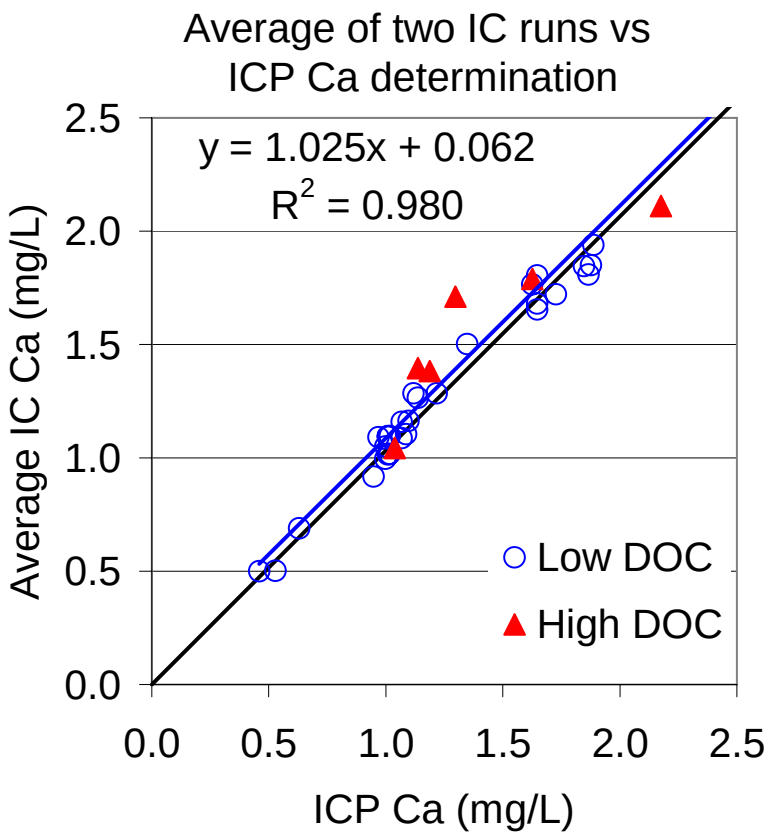
Newbert-E Dup	28-Jul
Newbert-O	27-Apr
Abol-O	29-Apr
Abol-E	12-Aug
Baker	3-Jun
Round-E Dup	22-Jul
Rocks HELM	14-Oct
Partridge-H	11-Aug
Upper Sister Lake	19-Aug
Penobscot	28-Oct
Hodge Pond TIME	27-Aug
Wiley-E RLTM	20-Oct

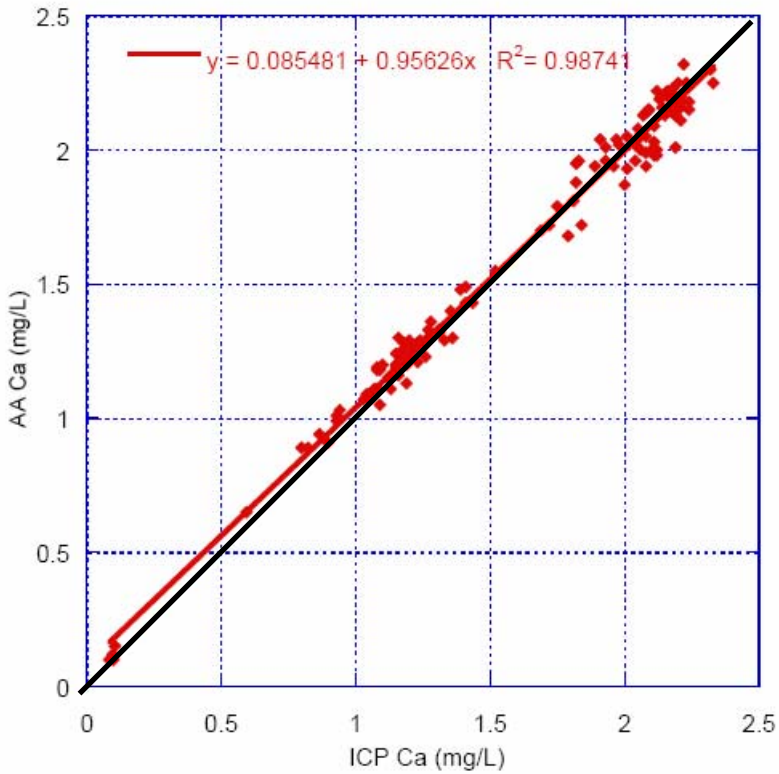
Calcium analysis
Comparison to ICP and IC reproducibility



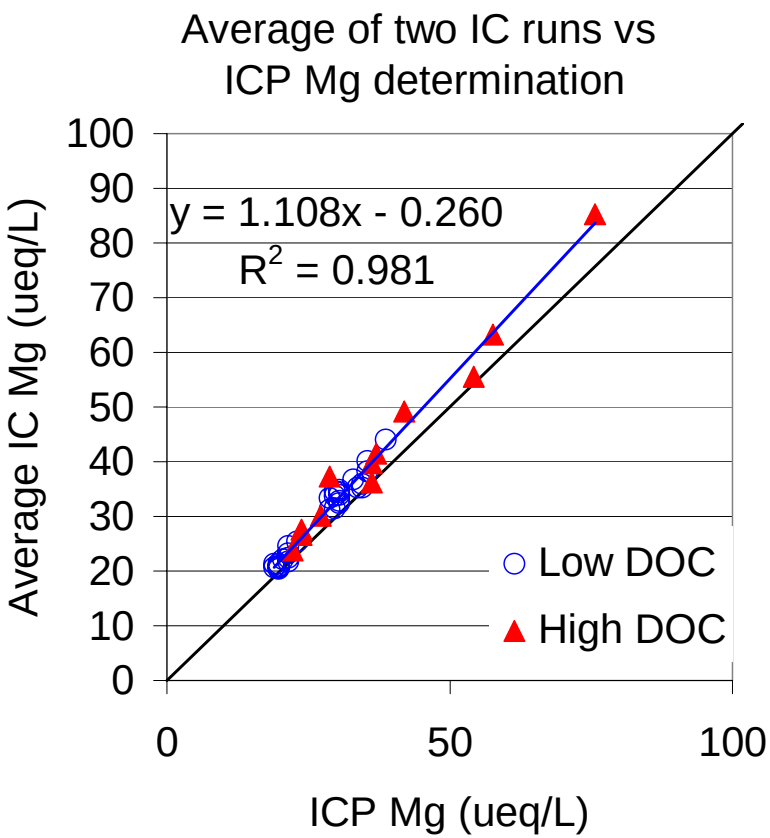


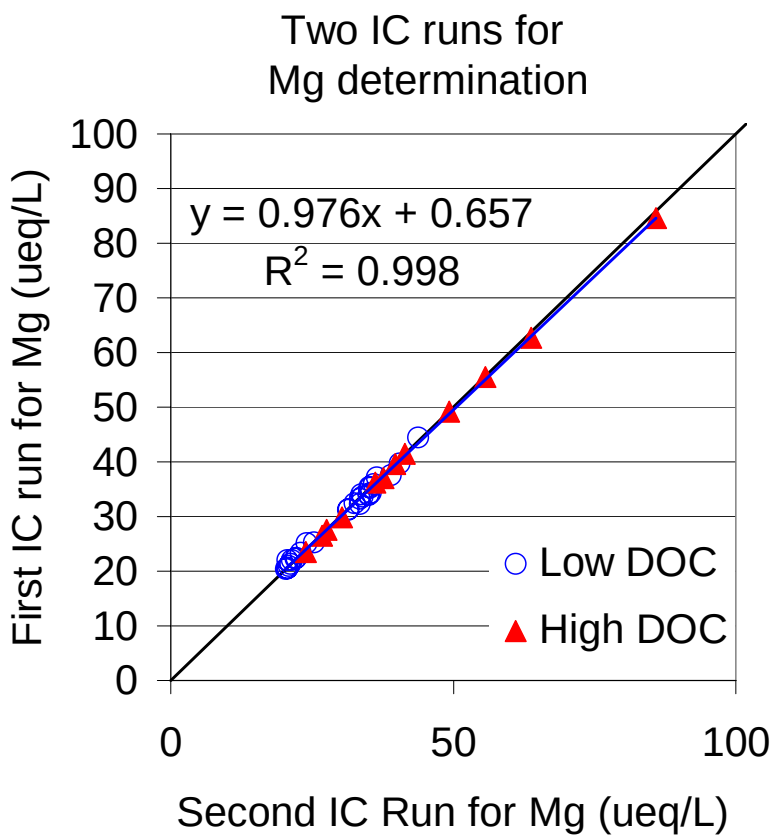
Calcium analysis
IC to ICP comparison vs AA to ICP comparison



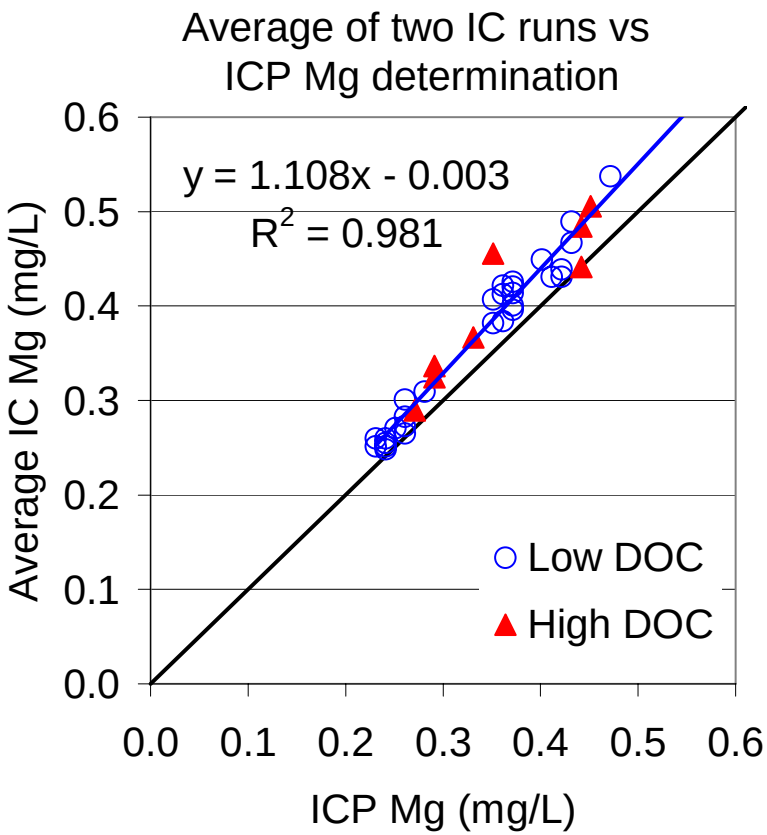


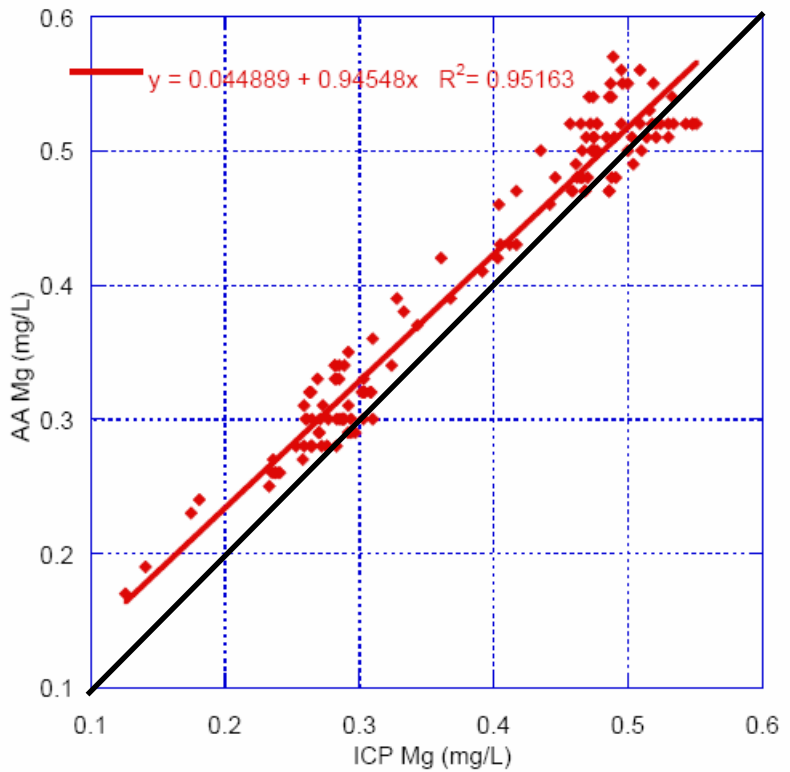
Magnesium analysis
Comparison to ICP and IC reproducibility



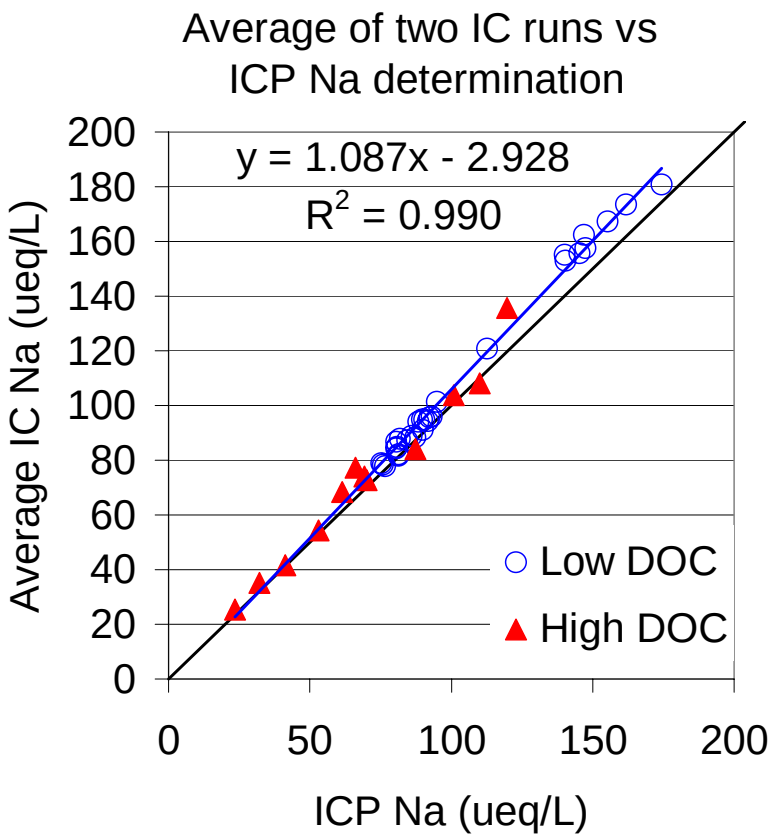


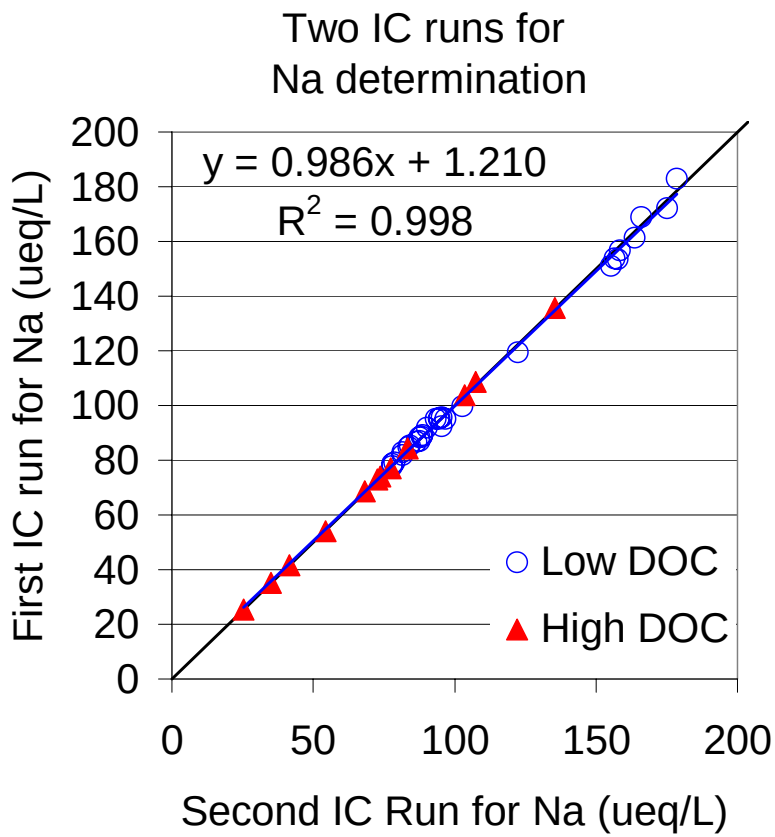
Magnesium analysis
IC to ICP comparison vs AA to ICP comparison



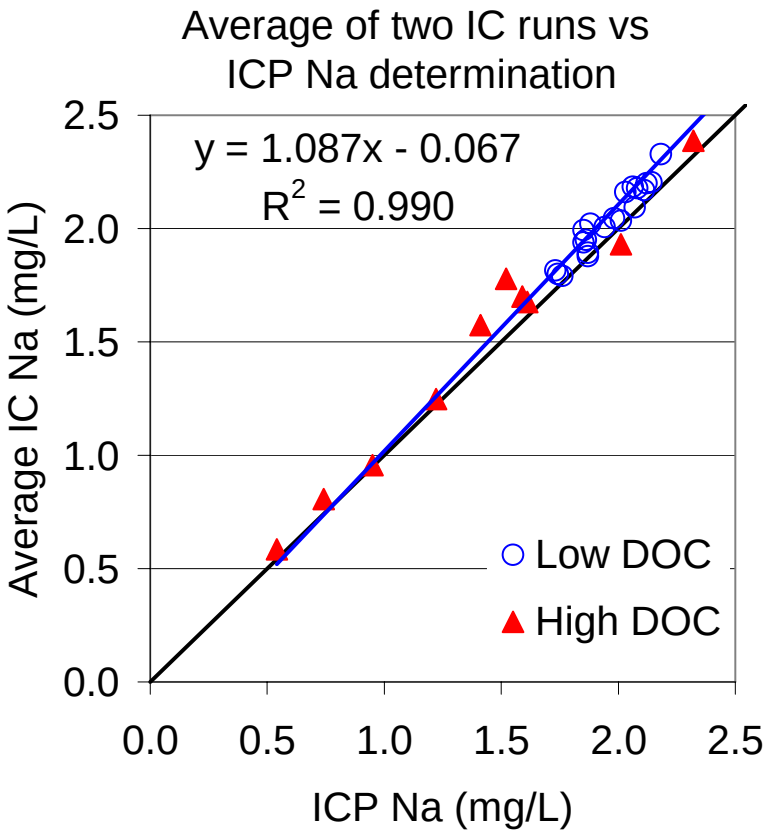


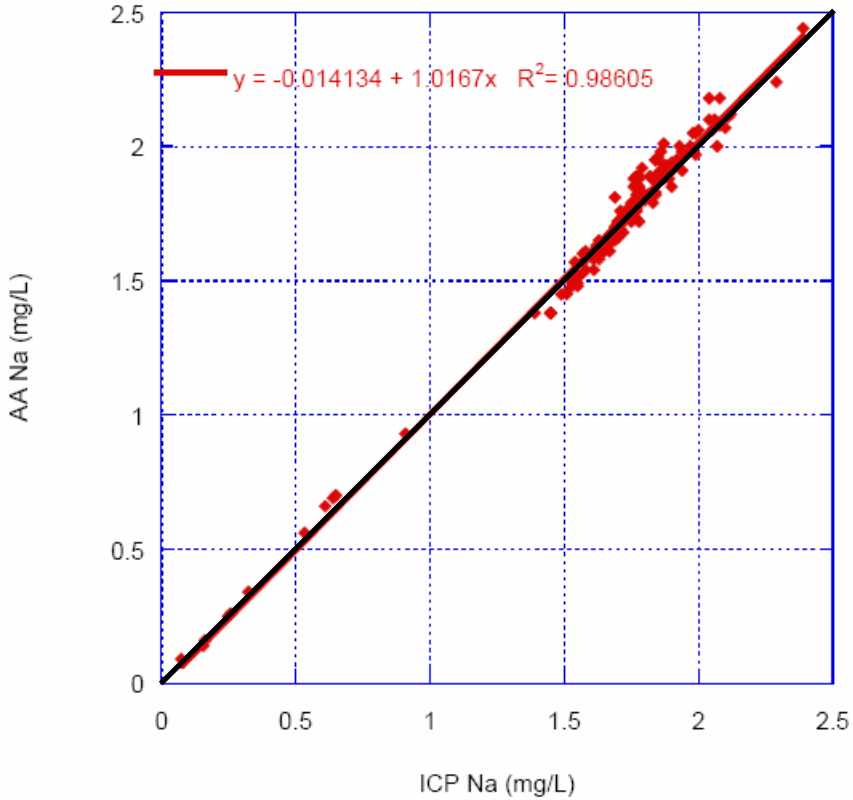
Sodium analysis
Comparison to ICP and IC reproducibility



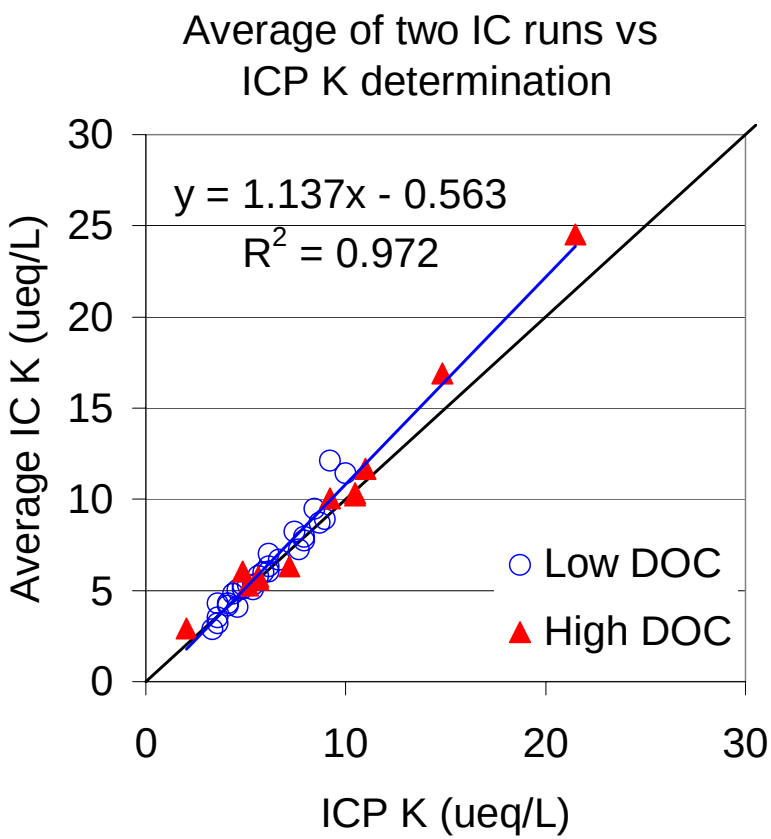


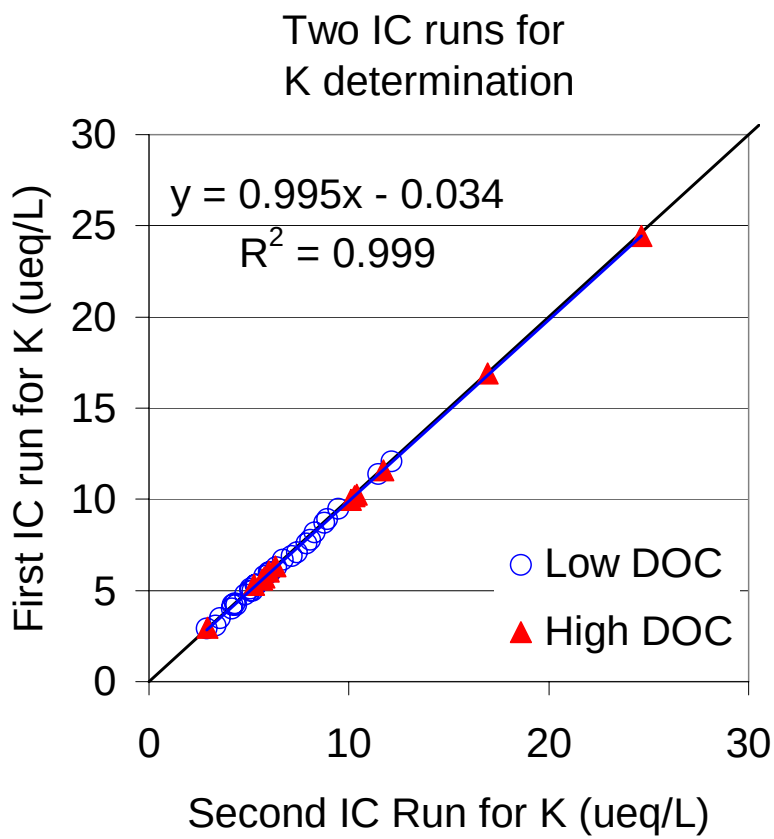
Sodium analysis
IC to ICP comparison vs AA to ICP comparison



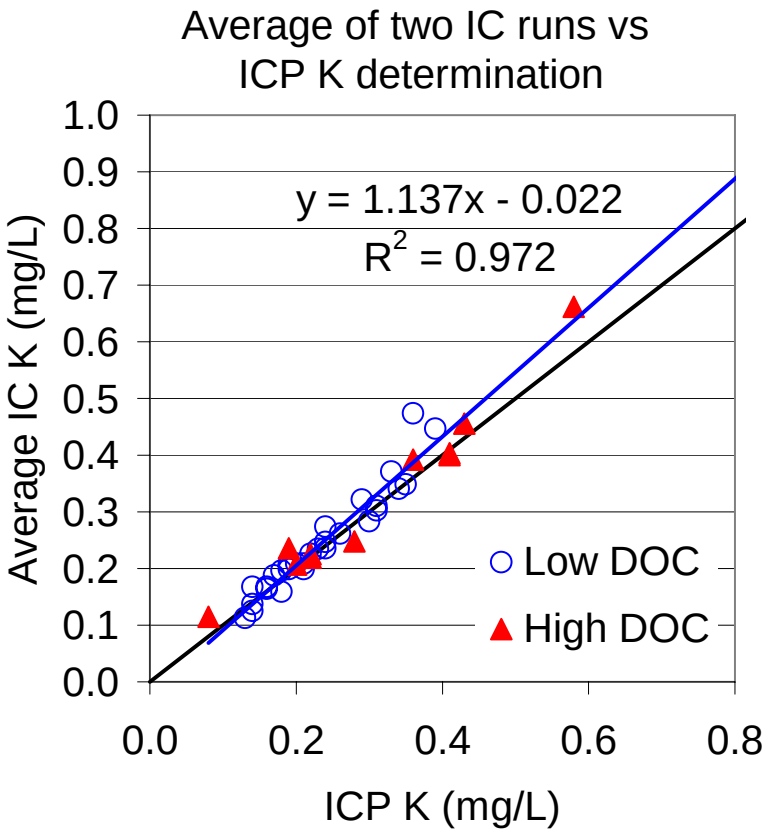


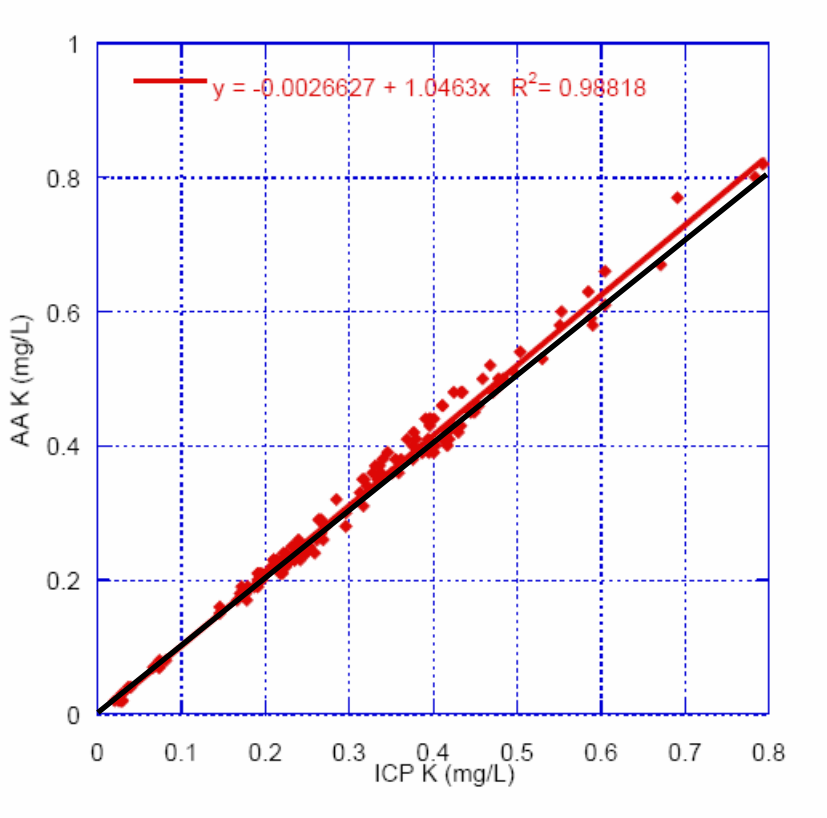
Potassium analysis
Comparison to ICP and IC reproducibility





Potassium analysis
IC to ICP comparison vs AA to ICP comparison





Summary Statements

	Table of r^2 values for comparisons			
	Ca	**Mg**	**Na**	**K**
IC Precision	0.999	0.998	0.998	0.999
IC vs ICP correlation (bias towards)	0.980 (IC)	0.981 (IC)	0.990 (IC)	0.972 (IC)
AA vs ICP correlation (bias towards)	0.987 (AA)	0.952 (AA)	0.986 (none)	0.988 (AA)

- Analyses were biased high towards IC for all comparisons
- Analyses were also biased high towards AA for all ICP vs AA comparisons except Na
- When data were analyzed separately, IC analyses differed from ICP analyses less among low DOC samples.
- NH_4 analysis is not running well yet. NH_4 may degrade too quickly because we do not acidify samples.

- Thanks to everyone for their help

Tanya – analysis, data management, and sample prep

Catherine – analysis and sample prep

Lisa – sample prep and IC setup

Ken – analysis, data management, and sample prep

SK – ideas, consulting, equipment procurement

FOREST LITTER MERCURY DYNAMICS AT ACADIA NATIONAL PARK

Executive Summary

Mercury deposition to the landscape is typically evaluated using data from precipitation Hg deposition. Litterfall is also believed to be a major flux of Hg to soils in forested landscapes, yet much less is known about litterfall Hg flux. To quantify this component of Hg cycling in forested landscapes, we measured litterfall Hg contributions to total Hg deposition in two small research watersheds in Acadia National Park, Maine. Notable findings of this research include:

1. The estimated annual deposition of Hg via litterfall in Hadlock Brook watershed (10.1 $\mu g\ m^{-2}$) and Cadillac Brook watershed (10.0 $\mu g\ m^{-2}$) was greater than precipitation Hg deposition and similar to or greater than the magnitude of Hg deposition via throughfall. These results demonstrate that litterfall Hg flux to forested landscapes is at least as important as precipitation Hg inputs.
2. The mean litter Hg *concentration* in softwoods (58.8 ± 3.3 ng Hg g^{-1}) was significantly greater than in mixed (41.7 ± 2.8 ng Hg g^{-1}) and scrub (40.6 ± 2.7 ng Hg g^{-1}), and significantly lower than in hardwoods (31.6 ± 2.6 ng Hg g^{-1}). In contrast, the mean weighted litter Hg *flux* was not significantly different among vegetation classes.
3. Landscape characteristics (i.e., aspect, elevation and canopy density) were significantly correlated with litter Hg concentrations and flux.
4. A significant negative correlation was defined between litter C:N and litter Hg concentrations suggesting that C:N could be a potential predictor of litter Hg concentrations.
5. In decomposing litter, Hg was strongly bound to the non-labile components of organic matter.

Introduction

Since terrestrial ecosystems take up a greater proportion of the earth's surface and have more complex atmospheric boundaries than freshwater ecosystems, they intercept more Hg from the atmosphere. The three predominant Hg input vectors to terrestrial ecosystems are precipitation, throughfall, and litterfall. Precipitation is any form of wet deposition that, can deliver Hg directly from the atmosphere to the landscape. Throughfall, precipitation that leaches through the vegetative canopy, includes wet and

dry deposition and foliar leachate. Litterfall consists primarily of leaves, twigs, bark, cones, seeds and other vegetative debris. The flux of Hg from litter is considered dry deposition since the atmosphere, rather than the soil, is the predominant source of Hg in foliage (Hanson et al., 1995; Lindberg, 1996; Bishop et al., 1998; Rea et al., 2002; Ericksen et al., 2003). Results from field research at forested sites indicate that Hg in precipitation is the smallest of the three fluxes, while litterfall is often the largest (St. Louis et al., 2001; Grigal, 2002; Miller et al., 2005).

The Mercury Deposition Network (MDN) currently monitors Hg in precipitation using sampling field stations distributed throughout the United States and Canada (Schroeder and Munthe, 1998). Acadia National Park (ANP) is one site within the MDN network that monitors Hg in precipitation on a continual basis. Since MDN only measures Hg deposition in precipitation, it is not possible to use MDN data to quantify spatial and temporal trends in *total* Hg deposition, nor provide suitable Hg input data for effective ecosystem Hg mass balance calculations. The quantification of the relative magnitudes of dry-deposition fluxes, such as throughfall and litterfall, has been identified as a gap in our knowledge of Hg in terrestrial ecosystems (Iverfeldt et al., 1996, Mason et al., 2005).

The main objective of this study was to quantify the magnitude of the litter Hg flux in forested watersheds in ANP. By doing this we can combine data on litter Hg with other Hg research results from these watersheds to determine the importance of litter Hg flux in Hg cycling. This report summarizes the important aspects of the topic of this research, namely Hg in litter. The following sections vary in format and length and are arranged to address the following questions:

1. What is the total annual deposition of Hg via litterfall in these watersheds?

2. What is the role of vegetation type in litter Hg dynamics?

3. Do aspect, elevation, and canopy cover correlate to Hg in litterfall?

4. Does litter C:N, as a measure of litter quality, influence Hg concentrations?

5. How does litter decomposition and leaching alter litter Hg concentrations?

Methods

Litterfall was sampled at a total of 19 sites in Hadlock Brook watershed and 20 sites in Cadillac Brook watershed (Figure 1). The network of 39 sites consisted of: (1) 12 core sites used for long-term sampling of litter, throughfall, and soil (Amirbahman et al., 2004; Johnson, 2002; Nelson, 2002) for the Park Research and Intensive Monitoring of Ecosystems Network (PRIMENet) study, (2) 17 throughfall study sites located along transects from ongoing throughfall studies (Nelson, 2002), and (3) 10 additional sites randomly located within under-represented vegetation types. Some of the throughfall transect study sites were omitted because they either over-represented a vegetation type, or were not located beneath a vegetative canopy. The study sites chosen provided both a balance of replication within major vegetation types to the extent possible while linking this research to other ongoing studies in these watersheds (Figure 1). At each site measurements of aspect, canopy cover, and elevation were conducted as described by Sheehan (2005).

Litterfall was collected in polyethylene basins, and unsorted dried samples were analyzed for total Hg, using a modification of EPA method 245.6 (*Determination of Mercury in Tissue*), by cold vapor atomic absorption (Sheehan, 2005). A sub-set of

samples were analyzed for total-C and total-N with a LECO CN Analyzer (Sheehan, 2005).

Differences among means by vegetation type were assessed for statistical significance by Analysis of Variance (ANOVA) at a 95% confidence level. Continuous data sets (elevation, canopy cover, and openness) were tested for significance of correlations with litterfall, litter Hg concentration, and Hg flux in litter. A modified ANOVA, utilizing contrasts, was used to test for interactions among landscape factors whenever possible. All means are reported with their corresponding standard error (± SE). Time-weighted mean litterfall and mean litter Hg flux values are reported to account for differences in the length of the collection periods used in this study.

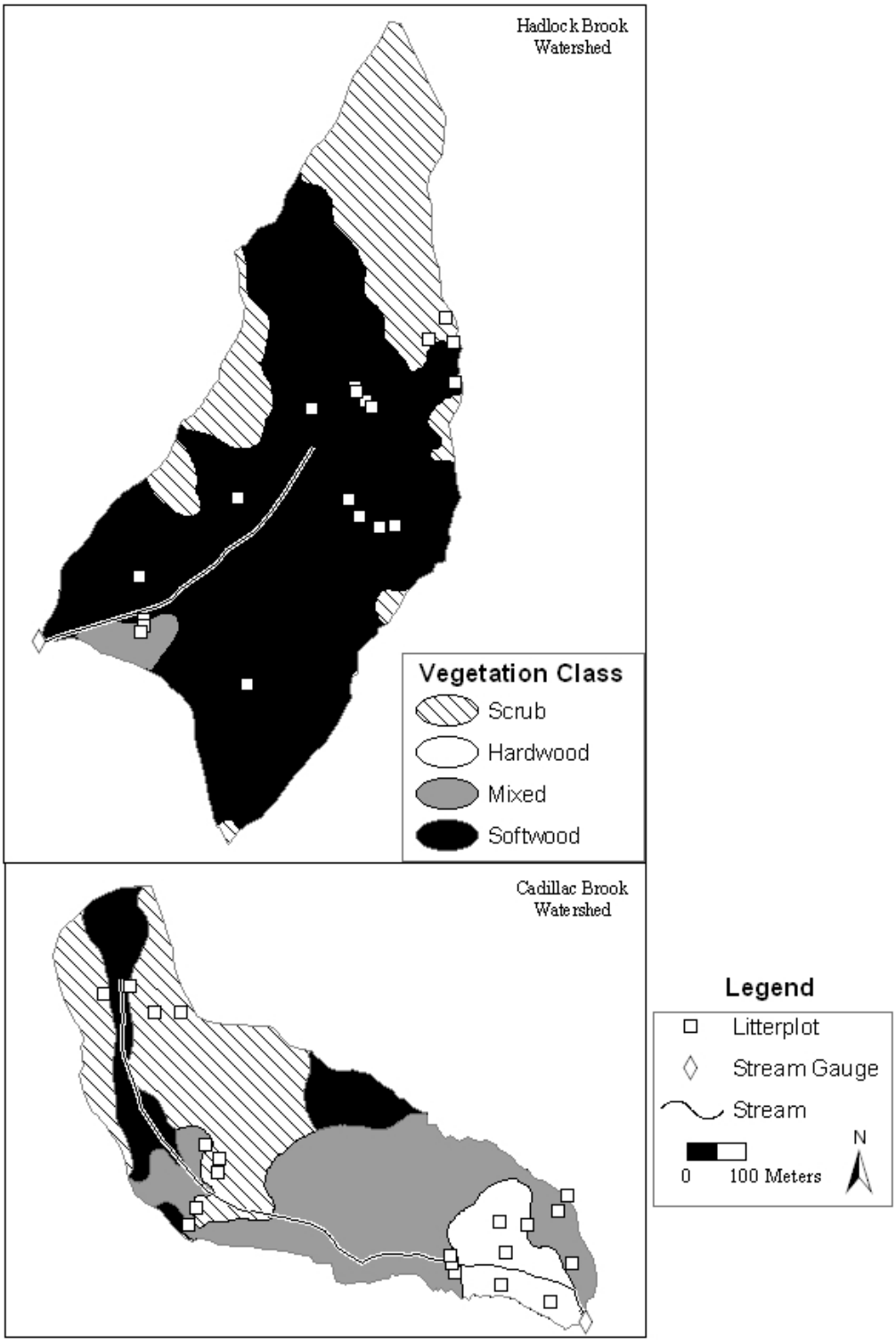


Figure 1. Map of vegetation type distributions and litter sample site locations within the study watersheds, Acadia National Park, Maine.

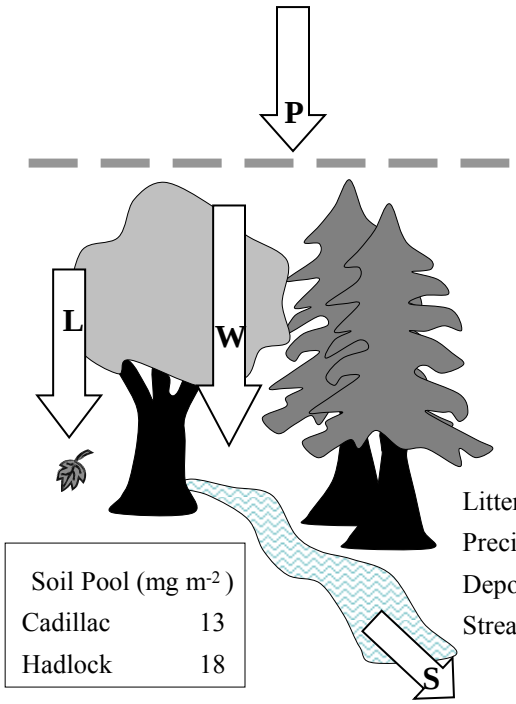
Results and Discussion

1. What is the total annual deposition of Hg via litterfall in these watersheds?

Figure 2 shows an estimated annual flux of Hg in litter for this study at ANP. The flux estimate was considered conservative and based on the study period as described in the methods. Also shown in Figure 2 are other available estimates of Hg data for these watersheds that include inputs, stream export, and soil pools. The overall estimated annual mean litter Hg flux across both watersheds was 10 $\mu g\ m^{-2}$. The annual estimates of Hg deposition in litter for each watershed were similar. The continuous input of litter in softwoods throughout the year balanced the uniquely large event of litterfall that occurred in hardwoods at the end of the growing season.

Although slightly lower, the annual Hg litter flux in ANP is similar to several relevant studies for forested ecosystems. Rea et al. (2002) reported the annual Hg litter flux was 11.4 $\mu g\ m^{-2}$ for the Lake Huron Watershed, MI, composed of northern hardwoods. Similarly, St. Louis et al. (2001) calculated an annual litter Hg flux of 12.0 $\mu g\ m^{-2}$ in the Experimental Lakes Area in Ontario, Canada, a northern boreal forest dominated by softwood species. Rea et al. (1996) estimated an annual litter Hg flux of 13.0$\mu g\ m^{-2}$ from 1994 data in a northern hardwood forest from the Lake Champlain Watershed, VT. For the same study site, Rea et al. (2002) determined that the annual flux of Hg in litter was 15.8 $\mu g\ m^{-2}$, from samples collected in 1995. The ANP annual Hg litter flux reported here was substantially lower than the 30.0 $\mu g\ m^{-2}$ determined by Lindberg (1996) in a temperate hardwood forest in TN, which unlike ANP, is located near known point sources of Hg. Annual Hg litter fluxes in Scandinavia reported by Lee

et al. (1998, 2000), were in the range of 18-60 μg m^{-2}, substantially higher than ANP. Historically, atmospheric Hg concentrations in those study areas were higher than those documented in the United States, which could explain why Scandinavian litter Hg fluxes were larger than in ANP (Lindberg, 1996; Grigal, 2002).



	Annual Fluxes (ug m^{-2})	
	Cadillac Brook	Hadlock Brook
Litter (L)	10.0	10.1
Precipitation (P)	6.8	6.8
Deposition (W)*	9.4	10.2
Streams (S)	0.4	1.3

Figure 2. Estimates of annual Hg flux and soil pools in the study watersheds. Precipitation flux (P) for the period of this litterfall study was measured at the ANP Mercury Deposition Network site ME98 (National Atmospheric Deposition Program, 2005). The Hg soil pool and Hg stream flux (S) were measured at these sites July 1999-2000 (Johnson, 2002; Amirbahman et al., 2004). Deposition (W)* is defined here as an estimate of annual wet deposition that included the summation of throughfall measurements from May 2000-November 2000 plus wet-only precipitation for the period November 1999-April 2000 as reported by Johnson (2002).

The ratio of mean annual Hg flux in precipitation (P) to Hg deposition in throughfall (W being the approximation used here) to litter Hg flux (L) is 1:1.5:1.5 overall for the study watersheds. Similar ratios were reported by Grigal (2002), 1:1.8:2.2,

and Munthe et al. (1995), 1:1.5:3. These data show that only monitoring Hg in precipitation can be misleading, since total Hg deposition is far greater.

The estimated annual flux of Hg in litter for each study watershed was well in excess of precipitation inputs, and similar to the deposition of Hg (W), calculated to include potential dry deposition of Hg as measured in throughfall during the field season and precipitation Hg inputs (as reported by MDN) for the remainder of the year. Dry deposition was not included in the deposition (W) figure for November1999-April 2000, and we therefore assume this estimate to be conservative.

Since annual Hg export via stream water was an order of magnitude smaller than the deposition estimates reported here (L, P, and W), it is reasonable to assume that either (a) the average soil pool of 16 mg m^{-2} is increasing, (b) an important export vector such as Hg volatilization was not measured and is significant, or (c) both of these are true. If we estimate from Figure 3.5 that total Hg deposition is ~20 μg $m^{-2}yr^{-1}$ (L+W), and export in streams is ~0.8 μg $m^{-2}yr^{-1}$, then an average soil pool of 16 mg m^{-2} for these watersheds represents ~833 years of deposition to the site if all inputs are retained. This gives a conceptual estimate of accumulation rates, but neglects important factors that could include the accuracy of our estimates based on limited data, the potential for larger soil pools given the limited sampling depths of Amirbahman et al. (2004), the lack of Hg volatilization data for these watersheds, the potential for changing rates of atmospheric Hg deposition, and disturbance. Indeed, Amirbahman et al. (2004) reported details of soil differences between these watersheds and noted the soil pool for Hadlock watershed was greater than the Cadillac watershed (Figure 2). They interpreted the lower Cadillac Brook soil pool as representing the effects of the fire of 1947 that affected only the

Cadillac Brook watershed, and the influence of a higher hardwood vegetative cover in Cadillac compared to Hadlock since that disturbance event.

Miller et al. (2005) estimated total annual Hg deposition, defined as the sum of Hg delivered via precipitation, dry aerosol deposition, litterfall, and cloud droplet assimilation, to be within the range of 20-25 μg m^{-2} for this study area using a GIS-based model. Their modeled Hg input estimates agreed well with our estimate of total annual Hg input of ~20 μg m^{-2} from this study.

2. What is the role of vegetation type in litter Hg dynamics?

In order to understand how litter Hg deposition varies across the landscape, it is important to identify the critical factors that influence Hg deposition flux. Differences in plant community composition are thought to be a critical site characteristic that governs litter Hg flux, and the following findings address the question of vegetation influences.

2.1. Litterfall Quantification

Collection periods varied from 30 to 133 days in length, and therefore litterfall mass for the individual collections are presented on a per day basis to allow for comparison among collections (Figure 3). The mean rate of litterfall normalized to a per day basis ranged from 0.10 ± 0.03 to 7.38 ± 1.9 g m^{-2} day^{-1}. The greatest variation among collectors within vegetation types for individual collections of litterfall occurred among hardwood sites, which produced the highest litterfall masses. The smallest variation occurred among scrub sites, which generally had the lowest litterfall due to the patchiness and stature of these vegetative communities. There was an axiomatic increase in litterfall in all vegetation classes during October and November and therefore our

collection program focused resources on this peak period. Litterfall in softwoods was more consistent throughout the year than hardwood sites. The high rates of litterfall in hardwoods in the autumn make hardwoods the overall highest litterfall vegetation type. Softwoods do not produce a major autumn litterfall event at the scale of hardwoods, but demonstrate modest yet consistent rates of litterfall throughout the winter which resulted in softwoods having the highest litterfall mass for the April 2004 collection (0.79 ± 0.03 g m^{-2} day^{-1}), representing the preceding winter. The litterfall variation for mixed vegetation sites was intermediate between hardwoods and softwoods among collectors for individual collections, and among collection periods over time.

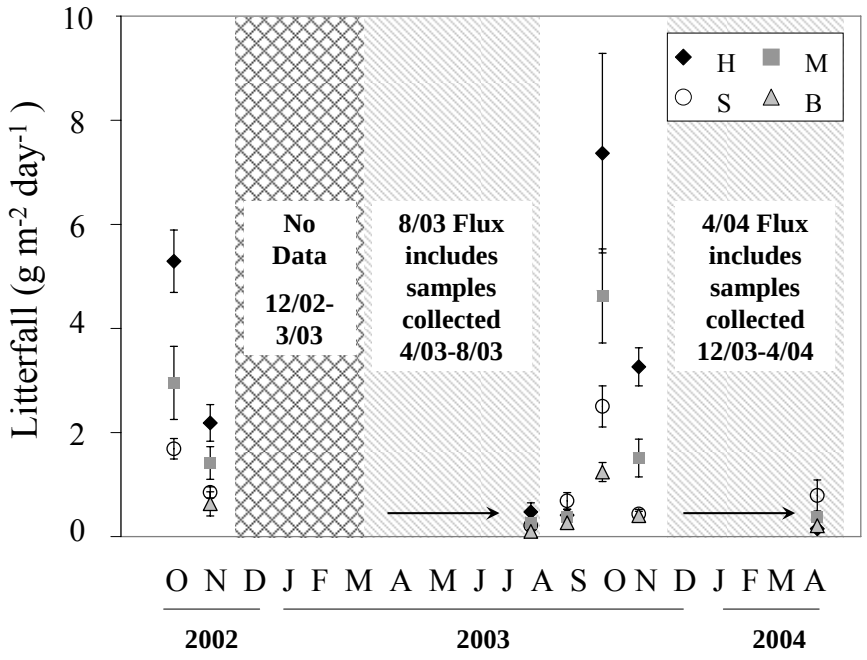


Figure 3. The mean and standard error for litterfall from each collection by vegetation class (H=Hardwood, S=Softwood, M=Mixed, B=Scrub) for the study period. Litterfall is expressed as g m^{-2} day^{-1} to account for varying collection periods.

When integrated over the entire study period the overall weighted means for litterfall were 1.55 ± 0.23, 0.87 ± 0.14, 0.78 ± 0.12, and 0.36 ± 0.05 g m^{-2} day^{-1} for

hardwoods, mixed, softwoods, and scrub, respectively. Although weighted mean litterfall mass for hardwoods was greatest, the difference between hardwoods, softwoods and mixed was not statistically significant. The weighted mean litterfall mass for the scrub vegetation type was significantly lower than the other vegetation types.

2.2. Hg Concentration of Litter

Descriptive statistics for the concentration of Hg in litter from all samples measured during this study are presented in Table 1. Figure 4A shows mean litter Hg concentrations from this study by vegetation type. Softwood sites had significantly

Table 1. The mean concentration (ng g^{-1}) of Hg in litter for all samples collected from October 2002-November 2003.

Mean	Std. Error	Median	Minimum	Maximum	n
46.9	1.9	41.3	10.7	133.4	153

higher mean litter Hg concentration (58.8 ± 3.3 ng Hg g^{-1}) than all other vegetation types. Presumably higher concentrations of Hg in softwood litter was primarily attributed to the duration of foliar exposure prior to leaf-fall and higher surface area for softwoods compared to hardwoods (Rassmussen et al., 1991; Rasmussen, 1995; Kolka et al., 1999; St. Louis et al. 2001; Grigal, 2003). Litter collected at hardwood sites had significantly lower Hg concentrations (31.6 ± 2.6 ng Hg g^{-1}) than any of the other vegetation types, consistent with the literature.

Mean litter Hg concentrations were not significantly different from each other for mixed and scrub vegetation types, but were significantly higher than hardwoods. Higher mean litter Hg concentrations in mixed and scrub vegetation types compared to hardwoods was likely due to the presence of litter from softwood species which were present at both mixed and scrub sites. Rasmussen et al. (1991) suggested that lower

stature plants may absorb Hg vapor emitted from soils, but it is unlikely that this explains the higher mean litter Hg concentration at scrub sites in this study since the soil in those vegetative units is thin and sometimes patchy.

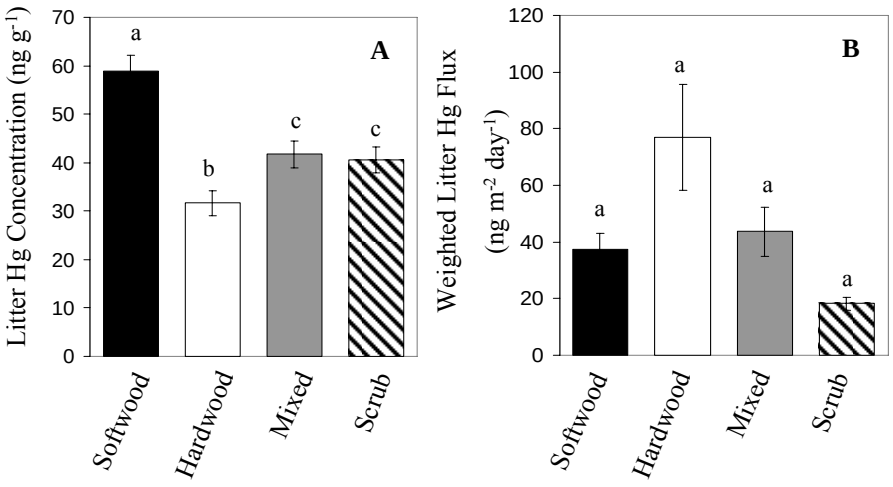


Figure 4. Mean (±SE) litter Hg concentrations (A) and Hg flux from litter (B) for each vegetation class in the study watersheds. Bars with the same lower case letter are not significantly different from each other at $p<0.05$.

Results reported here for ANP hardwood litter Hg concentrations were similar to those in a study of Michigan hardwood litter, but somewhat lower than the mean concentration of Hg in Vermont and Tennessee hardwood litter (Table 2). The mean litter Hg concentration at softwood sites in ANP is similar to other softwood literature values (Table 2). Previous research that measured litter Hg concentrations in these ANP study watersheds using a limited number of samples (n=10) reported a similar but smaller range (Johnson, 2002).

Table 2. Litter Hg concentrations in different vegetation types at various study locales.

Location	ng g^{-1}	Vegetation Type	Source
Tennessee, USA	105	Hardwood	Lindberg (1996)
Tennessee, USA	61	Softwood	Lindberg (1996)
Ontario, Canada	33-79	Jack Pine	St. Louis et al. (2001)
Ontario, Canada	25-30	Jack Pine/Birch	St. Louis et al. (2001)
Vermont, USA	47	Mixed Hardwood	Rea et al. (2002)
Michigan, USA	33	Mixed Hardwood	Rea et al. (2002)
Maine, USA	40-51	Mixed	Johnson (2002)

2.3. Field Season Litter Hg Flux

The mean weighted flux of Hg in litter, during field season 2003, is presented in Figure 4B. There were no significant differences in litter Hg flux among vegetation classes ($p<0.05$, Figure 4B), despite the range in mean weighted litter Hg fluxes and the significant differences evident in the concentration of Hg in litter (Figure 4A). The lack of significant differences in litter Hg flux was attributable to both the high variability in these data (particularly for hardwoods), the composite nature of the samples, and the inverse relationship between Hg concentration and litterfall mass when comparing hardwoods and softwoods. St. Louis et al. (2001) demonstrated that the relatively low mass of litterfall offsets the effectiveness of softwood Hg scavenging when comparing hardwoods and softwoods. They reported results from a softwood site with high Hg concentration but a low annual flux of Hg in litter, while litter from a hardwood site with a lower Hg concentration had a higher annual flux.

The Hg flux in litter was calculated for the 2003 field season (= 0.57 yr) by watershed on an area-weighted basis to account for the different spatial extents of each vegetation type in each watershed. The averaged Hg flux in litter was 7.5 μg m^{-2} for the growing season. The 2003 field season flux of Hg in litter was 8.7 μg m^{-2} in Cadillac

Brook watershed and 6.0 μg m^{-2} in Hadlock Brook watershed. The higher litter Hg flux in Cadillac reflected the greater spatial extent of hardwoods in Cadillac Brook watershed, which ultimately dominated these results due to the high hardwood litter flux in the latter part of the growing season. The high litterfall period in the autumn for hardwoods, and more consistent litterfall flux throughout the year for softwoods, result in important differences between these forest types for growing season, winter, and annual litter and litter Hg flux estimates.

3. Do aspect, elevation, and canopy cover correlate to Hg in litterfall?

Except for vegetation type, limited research has been conducted on ecosystem variables that affect Hg deposition (Mason et al., 2005). Therefore, our goal was to identify landscape characteristics that influence Hg assimilation or deposition. These characteristics should be easily measured to be most useful to researchers concerned with Hg deposition.

Air mass trajectories can dictate Hg source-sink relationships that govern deposition. Topography enhances Hg deposition when high relief areas stall the movement of air masses (Evers, 2005; Godbold, 1994; Jagels et al., 1989; Malcolm et al., 2003). Kittredge (1973) notes that forest canopies slow the velocity of wind. When air flow is decreased, foliar Hg uptake may be optimized. In a study of Hg in throughfall, Iverfeldt (1991) attributed depositional disparities to watershed orientation. Therefore the goal of this study was to determine if landscape characteristics, specifically elevation, aspect and canopy density, were correlated with litter Hg concentration or litter Hg flux. Correlations, although not predictive, can be used to indicate whether relationships warrant investigation because of parallel trends in the data between litterfall Hg and

easily measured landscape characteristics. The results reported here can be considered the first in a multi-stage process with the long-term goal being the identification of landscape characteristics that can be used to predict Hg deposition.

3.1. Aspect

The greatest weighted mean litterfall was found at sites facing southeast (Table 3). Sites facing southwest exhibited a significantly lower amount of litterfall than those facing southeast, while litterfall at northwest facing sites were not significantly different from any of the other aspect classes. One interpretation of these results could be that southwest facing sites were most exposed to harsh prevailing winds coming up the coast resulting in thinner canopies with less biomass to contribute.

Table 3. Mean (±SE) litterfall, litter Hg concentration, and litter Hg flux by aspect. Means followed by the same lowercase letter are not significantly different.

Aspect	Weighted Litterfall ($g\ m^{-2}\ d^{-1}$)	Hg Concentration ($ng\ g^{-1}$)	Weighted Hg Flux ($ng\ m^{-2}\ d^{-1}$)
NW	0.76a,b ± 0.16	53.5a ± 3.9	45.8a ± 10.0
SW	0.62a ± 0.13	55.3a ± 3.5	24.5a ± 3.5
SE	1.10b ± 0.15	37.9b ± 2.2	56.0a ± 9.4

For the period January 2003-May 2004, at ANP, the mean hourly wind direction was 196º (southwest as defined here) and the hourly wind speed ($m\ s^{-1}$) ranged from ~0.5 to ~16.1, with a grand mean of 3.4 (Acadia National Park, unpublished data). Wind data do not indicate extreme conditions, yet other climatic conditions or pollution may cause mean canopy cover to be lowest at sites facing southwest (60.6%) compared to sites facing northwest (85.3%) and southeast (69.6%), partially substantiating this interpretation. Additionally the wind data is recorded at McFarland Hill Air Research

Station at ANP, which is located slightly further inland than the study watersheds, and may be more protected from direct exposure.

Despite wind characteristics and canopy biomass, our observations suggest that among aspect classes, litterfall is dominated by more than just aspect, and that the composition of vegetation covaries with aspect and likely dominates litterfall results reported here. Much of the lower reaches of Cadillac Brook watershed, which faces predominantly southeast, were dominated by hardwood species that contribute greater amounts of litterfall than softwood communities. The southwest facing sites in Hadlock Brook were dominated by softwood species that had lower litterfall rates for the study period than hardwoods.

The mean concentration of Hg in litter from southeast-facing sites was significantly lower than any of the west-facing sites (Table 3). The southwest and northwest facing sites may be better situated on the landscape to intercept Hg enriched air masses originating from the west, as is the case for other sites in the Northeast (Evers, 2005; Malcolm et al., 2003). However, prevailing winds, although classified as southwest in the context of this study, are predominantly off the ocean at 196º, almost due south. A test of contrasts within ANOVA showed a significant ($p<0.10$) interaction between vegetation type and aspect. This, however, suggests that mean litter Hg concentrations in different aspect classes of these data could be partially explained by vegetative differences, since no hardwood sites faced northwest or southwest, and only 13% of the sites facing southeast were softwood sites. In light of the confounding interactions between vegetation and aspect in this study, we conclude that vegetation type was the predominant influence on litter Hg concentrations.

The highest mean weighted litterfall and the lowest mean litter Hg concentrations were found at southeast sites, while the opposite was true for both west facing classes. As a result, there were no significant differences in mean weighted litter Hg flux among aspect classes (Table 3).

3.2. Elevation

Results indicated a weak but significant negative correlation between elevation and litterfall, and a positive but weak correlation with Hg concentration over the limited range of elevation sampled, ~350 m (Table 4). Less litterfall at higher elevations may be an artifact of vegetation, rather than elevation directly, since community composition changes with elevation. Earlier we showed that scrub sites, which were located at the higher elevations in the watersheds, have significantly less litterfall than other vegetation types that occupy lower elevations in the watersheds. As elevation increases the amount of softwood litter contribution also increases.

Table 4. Correlation coefficients (r) for landscape characteristics and litterfall, litter Hg concentration, and litter Hg flux. Superscripts denote significance at $p<0.05$ (**) and $p<0.10$ (*).

	Elevation	Canopy Cover	Openness
Litterfall	-0.34^{**}	0.25^{**}	-0.18
Hg Concentration	0.17^{**}	0.15^{*}	-0.20^{**}
Hg Flux	-0.25^{*}	0.26^{*}	-0.22

Lindberg (1996) suggested that the presence of moisture enhances the assimilation of Hg into foliage so coastal fog and clouds, which regularly occur in ANP, may increase Hg concentrations at both low and high elevations. Perhaps increasing Hg concentrations at higher elevations, as indicated by these results, are controlled by a stronger moisture effect near the summits, since cloud water has a greater liquid water

content than fog (Malcolm and Keeler, 2002; Miller et al., 2005). Also, vegetation at higher elevations may accumulate more Hg from the atmosphere, due to an orographic mechanism allowing vegetation to assimilate Hg (Malcolm et al., 2003; Evers 2005). As presented in section 2, scrub sites, located at high elevations in this study, did not show greater concentrations of Hg than in vegetation situated lower in the landscape. It is likely that elevation reflects meteorological and edaphic factors that strongly influence the distribution and character of vegetation on the landscape, and it is the vegetation that has the most direct effect on Hg scavenging, and subsequently litter Hg concentration.

Litter Hg flux was negatively correlated with site elevation reflecting a stronger negative litter correlation than positive Hg concentration correlation (Table 4). No significant interaction between vegetation type and elevation was found.

3.3. Canopy Density

Theoretically, openness is the inverse of canopy cover, but both metrics provide loose approximations of canopy density, or vegetative biomass, at sampling sites. The strongest correlation for canopy cover was a positive and significant correlation with litterfall, which is logical since it is the canopy that produces the litter collected (Table 4). Although we expected there to be a significant correlation between openness and litterfall, there was none. Had openness been measured at all 39 sampling sites, rather than 29, the correlation may have been more parallel to canopy cover.

There was a weak, but significant, positive correlation between canopy cover and litter Hg concentration, and a stronger negative correlation with openness. Although there is a higher surface area in softwood foliage compared to other vegetation types, which leads to a higher scavenging efficiency for softwood vegetation (Rasmussen et al.

1991; Kolka et al., 1999; St Louis et al., 2001), the correlations presented here were not demonstrably the result of canopy density differences between vegetation types. Mean openness at hardwoods sites (73.8%) was greater than mean openness at softwood sites (57.5%). Mean canopy cover values showed the opposite relationship, with the canopy being slightly denser at hardwood sites (mean cover, 78%) than at softwood sites (mean cover, 76.5%). Regardless of vegetation type-based generalizations, taken together these correlations suggest that denser canopies retard air movement, which was discussed by Kittredge (1973), and allow for greater Hg uptake efficiency, not unlike topography.

There was a weak positive correlation between canopy cover and Hg litter flux ($p=0.10$). Since foliar biomass drives litter Hg flux, we expected canopy cover to be significantly correlated to litter Hg flux (St. Louis et al., 2001). The small sample sizes and variability in the data in this project may explain why this correlation was not stronger, and no significant correlation was evident between Hg litter flux and openness.

3.4. Synthesis of Landscape Characteristics and Litterfall Mercury

We found aspect, elevation and canopy density to often be significantly correlated with litter Hg concentrations and flux although most correlations were relatively weak. The confounding effects of simultaneous differences in landscape characteristics and vegetation types obscured our ability to draw conclusions about direct linkages from this study. What does emerge from these analyses, however, is the predominant effect of vegetation characteristics on Hg dynamics in these extensively managed landscapes.

The proximity of ANP to the coast, and the fact that winds often originate from the open ocean to the south rather than west, likely also plays an important role in litter Hg deposition at this site. Since the flux of Hg emitted from oceans is estimated to be

half as much as anthropogenic emissions and two times the flux of Hg emitted from natural terrestrial sources (Schroeder and Munthe, 1998), the assumption that Hg pollutant inputs are dominated by the westerly winds could be erroneous. Also, since this study site has the greatest relief along the entire east coast, it is impacted by coastal fog and clouds. Cloud water Hg concentrations are often much higher than the concentration of Hg in precipitation (Malcolm et al., 2003; Miller et al., 2005). Although no known research has been conducted on the Hg concentration of coastal fog, which frequently shrouds the ANP landscape, Jagels et al. (1989) documented very acidic fog at ANP (3.3 pH), indicating the pervasiveness of some atmospheric pollutants. Research on Hg in coastal fog, and documentation of its frequency and spatial distribution, would provide useful insights to understand Hg cycling at ANP.

Deposition of Hg in litter at ANP likely reflects a combination of climatic influences, proximity to the ocean, topography, and vegetation. Evers (2005) indicated that mid-coast Maine is a biological hot-spot for Hg accumulation, attributed in part to a combination of landscape characteristics. Further study is required to understand the mechanisms of influence of these landscape characteristics.

4. Does litter C:N, as a measure of litter quality, influence Hg concentrations?

Litter quality, or the amount of nutrients available for decomposition, differs among vegetation types. The quality of organic matter in litter from softwoods, as characterized by high carbon (C) to nitrogen (N) ratios, is considered ecologically lower than litter organic matter quality from hardwoods with relatively low C:N (Delaney et al., 1996; Magill and Aber, 2000). The concentration of Hg is generally greater in softwood

litter relative to hardwoods (Grigal, 2002). The following pilot study was an investigation into the possible link between Hg, C, and N concentrations in litter collected in ANP.

There was a significant, negative correlation between litter Hg concentration and the litter C:N ratio (Figure 5). This outcome is contrary to our expectation that litter C:N would reflect differences in litter from different vegetation types, with a positive relationship between C:N and Hg concentration. A positive slope was expected if litter from hardwood sites had low Hg concentrations and low C:N, while litter from softwood sites had high Hg concentrations and high C:N. As shown in Figure 5, the sample site vegetation classification had no bearing on the relationship between litter C:N and Hg.

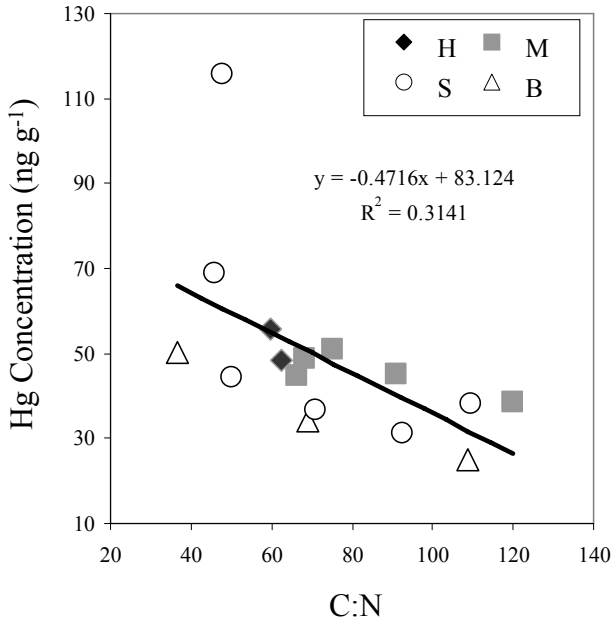


Figure 5. Relationship between litter Hg concentration and litter C:N ratio for samples from sites with different vegetation classes (H=Hardwood, S=Softwood, M=Mixed, B=Scrub).

The range of litter C:N in these data includes typical C:N values for litter and foliage in the lower end of the range, but the higher C:N values likely reflect the presence of woody tissue in the litter samples (e.g., branches and twigs). This is reasonable since woody tissue Hg concentrations are also lower than foliar/litter Hg concentrations (Rasmussen et al. 1991; Fleck et al., 1999; Grigal, 2003). Woody tissue also has low N concentrations, which would explain why we found a positive correlation ($r = 0.59$, $p<0.10$) between litter Hg concentration and litter N concentration. The objectives of this study were to evaluate bulk litterfall, and therefore litterfall components were not sorted giving our litter sample population a high degree of variability with respect to herbaceous versus wood tissue composition, as is evident in this C:N and Hg relationship.

While the relationship between litter C:N and litter Hg does not indicate a cause and effect relationship, these pilot data suggest that bulk litter C:N is a potentially useful predictor of Hg concentrations. At a minimum, C:N may be an important litter quality variable to consider when evaluating litter Hg dynamics.

5. How does litter decomposition and leaching alter litter Hg concentrations?

Litterfall is a major pathway for Hg deposition in forested systems. Little is known about the fate of litter Hg once it reaches the forest floor. Litter undergoes decomposition that is affected by moisture, temperature, and microbial activity (Melillo et al. 1982). Like many metals, Hg concentrations increase in litter over time as decomposition progresses (Tyler, 2005).

To explore the influence of time on litter Hg processes, we conducted a pilot study to assess litter Hg concentrations at various stages of exposure to field conditions.

One litterfall sample was collected from each watershed (Oct. 2003) and divided into five sub-samples (~10g each) composed of only hardwood species and subjected to three treatments. Treatment A involved immediately drying and analyzing the samples for total Hg. Treatment B involved burying litter samples in sealed polyethylene bags within the O horizon for the winter months (~5.5 months), then drying and analyzing the samples. Samples subjected to Treatment C were treated similarly to those in Treatment B, but leached in 200 ml of fresh rainwater in a sealed Teflon bottle for two weeks before total Hg analysis. Animal damage caused the loss of 60% of the sub-samples in Cadillac, but the full series was intact for Hadlock Brook watershed (HHA-HHE).

Despite sample loss, Figure 6 shows the results that were obtained from this study. The data suggest that litter Hg concentration increases over the winter months. This trend is most likely due to a decrease in overall litter mass through decomposition, with minimal loss of Hg. Since litter Hg concentrations were both higher (HHE) and lower (HHD & HHC) after leaching, compared to un-leached samples (HHB), leaching did not appear to have an important effect on Hg bound within the litter. The lack of any significant Hg loss due to decomposition of the litter mass as well as leaching suggests that Hg is strongly bound to the non-labile organic matter in the litter. The influence of field conditions, precipitation, and decomposition on Hg in litter warrants further study.

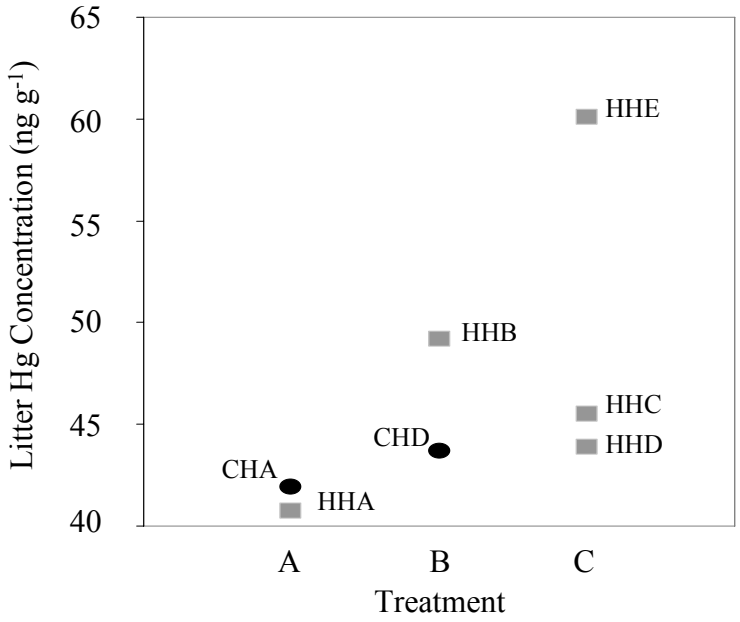


Figure 6. The concentration of Hg in litter subjected to three treatments (A= immediate total-Hg analysis, B= over-winter soil incubation followed by total-Hg analysis, and C= over-winter soil incubation, a two week precipitation leach, and then total-Hg analysis).

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